

(19)日本国特許庁 (JP)

(12) 公開特許公報 (A)

(11)特許出願公開番号

特開平9-143238

(43)公開日 平成9年(1997)6月3日

(51)Int.Cl. ⁶	識別記号	府内整理番号	F I	技術表示箇所
C 08 G 12/30	NCU		C 08 G 12/30	NCU
C 08 L 61/24	LNR		C 08 L 61/24	LNR
61/26			61/26	
61/28			61/28	

審査請求 未請求 請求項の数7 O L (全17頁)

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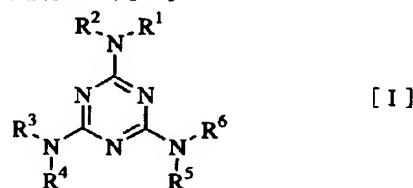
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(54)【発明の名称】 新規なアミノ樹脂組成物

(57)【要約】 (修正有)

【課題】 メラミン樹脂の耐熱性、高い硬度、すぐれた光沢などの特性を損なうことなく、強靭性、曲げ加工性、親油性を与えるアミノ樹脂組成物。

【解決手段】 式 [I]



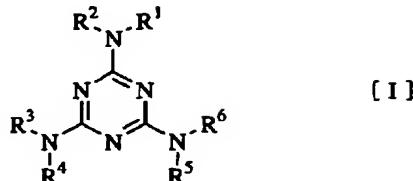
〔式中、置換基R¹～R⁶は各々互いに独立しており、それら置換基の1ないし5個は、C₁₋₂₀のアルキル基、C₂₋₂₀のアルケニル基またはフェニル基を表し、そして残りの置換基は水素原子を表す。〕により表されるトリアジン誘導体の1種または2種以上の混合物とホルムアルデヒドとを反応させることにより製造される新規なアミノ樹脂組成物；前記トリアジン誘導体の1種又は2種以上の混合物およびメラミン又は尿素の混合物にホルムアルデヒドと反応させることにより製造される新規なア

ミノ樹脂組成物、及びメラミン樹脂、メラミン・尿素-ホルムアルデヒド樹脂又は尿素ホルムアルデヒド樹脂からなるアミノ樹脂組成物。

【特許請求の範囲】

【請求項1】 式 [I]

【化1】

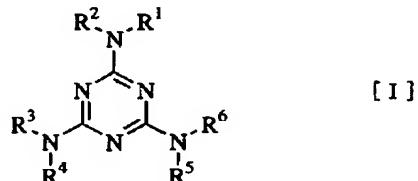


〔式中、置換基、R¹、R²、R³、R⁴、R⁵およびR⁶は各々互いに独立しており、それら置換基の1ないし5個は、C₁₋₂₀のアルキル基、C₂₋₂₀のアルケニル基（該アルキル基またはアルケニル基は、その構造中に任意に脂環式構造またはフェニル基を有していても良く、また2個の該アルキル基またはアルケニル基が同一窒素原子上にある場合はその窒素原子と一緒にになって3～8員の含窒素複素環を形成しても良い）またはフェニル基を表し、そして残りの置換基は水素原子を表す。〕により表されるトリアジン誘導体の1種または2種以上の混合物にホルムアルデヒドを反応させることにより製造されることを特徴とするアミノ樹脂組成物。

【請求項2】 式 [I] により表されるトリアジン誘導体の1種または2種以上の混合物1モルに対して1モルから該トリアジン誘導体の1種または2種以上の混合物のアミノ基の未置換水素原子と同数のモルのホルムアルデヒドを反応させることにより製造される請求項1記載のアミノ樹脂組成物。

【請求項3】 式 [I]

【化2】



〔式中、置換基、R¹、R²、R³、R⁴、R⁵およびR⁶は各々互いに独立しており、それら置換基の1ないし5個は、C₁₋₂₀のアルキル基、C₂₋₂₀のアルケニル基（該アルキル基またはアルケニル基は、その構造中に任意に脂環式構造またはフェニル基を有していても良く、また2個の該アルキル基またはアルケニル基が同一窒素原子上にある場合はその窒素原子と一緒にになって3～8員の含窒素複素環を形成しても良い）またはフェニル基を表し、そして残りの置換基は水素原子を表す。〕により表されるトリアジン誘導体の1種または2種以上の混合物およびメラミン又は尿素からなる混合物にホルムアルデヒドを反応させることにより製造されることを特徴とするアミノ樹脂組成物。

【請求項4】 請求項1に記載のアミノ樹脂組成物及びメラミン-ホルムアルデヒド樹脂、メラミン・尿素-ホ

ルムアルデヒド樹脂又は尿素ホルムアルデヒド樹脂を含有するアミノ樹脂組成物。

【請求項5】 置換基、R¹、R²、R³、R⁴、R⁵およびR⁶のうち、R¹、R³およびR⁵の1ないし3個がC₁₋₁₀のアルキル基を表し、残りの置換基が水素原子を表す請求項1ないし4のいずれかに記載のアミノ樹脂組成物。

【請求項6】 置換基、R¹、R²、R³、R⁴、R⁵およびR⁶のうち、R¹、R³およびR⁵の1ないし3個がメチル基、エチル基、n-ブロピル基、イソブロピル基、n-ブチル基、シクロヘキシル基又は2-エチルヘキシル基を表し、残りの置換基が水素原子を表す請求項5に記載のアミノ樹脂組成物。

【請求項7】 請求項1ないし6のいずれかに記載のアミノ樹脂組成物を含む化粧板、積層板、成形材料、塗料又は接着剤。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】 本発明は強靭性、耐熱性、耐光性、耐汚染性などにすぐれたアミノ樹脂組成物に関する。本発明組成物は成形材料、化粧材、塗料用硬化剤、木質系接着剤等に有用である。

【0002】

【従来の技術】 メラミン樹脂はそのすぐれた耐熱性、高い硬度、高い光沢、硬化速度が早いこと、成形時金型との離形性が良い事などの理由により従来成形材料、化粧材、塗料用硬化剤、木質系接着剤等に広く用いられている。

【0003】 一方、メラミンは6官能性である事、トリアジン骨核が剛直である事のためホルムアルデヒドとの樹脂化に際し、その硬化物は極めて架橋密度が高くなり、非常に硬く、耐熱性にすぐれる反面、もろく、その成形材料を工業用部品として用いる場合には制約があった。

【0004】 そのため成形材料として用いる場合、メラミン樹脂をゴムで変性したりフェノールで変性したりする試みがなされているが充分な効果を得るまでに至っていないのが、現状である。又各種アルコール、糖類などをメラミン樹脂組成物の中に加え耐クラック性、機械的強度を向上する事が行なわれているが、製造時共縮合が充分でないと経時に添加物がブリードアウトし、機械的強度の低下、耐クラック性の低下、表面光沢度低下などを引き起す欠点があった。

【0005】 またメラミン樹脂は主としてアルキッド樹脂の硬化剤として焼きつけ塗料に用いられるが、耐熱性、耐候性、耐スクラッチ性にすぐれる反面、塗膜の曲げ性、後加工性に劣り、そのためベンゾグアナミン、アセトグアナミン等のグアナミン類を用いる事があるが、ベンゾグアナミン樹脂はその構造の中にベンゼン基を有するため、耐光性が悪いことが知られており、使用用途

に制約がある。またメラミン-ホルムアルデヒド樹脂は有機溶媒に対する溶解性がなくそのため有機溶媒系塗料として用いる場合はメラミン-ホルムアルデヒド反応物をさらにメタノール、エタノール、プロパノール、イソプロパノール及びブタノール等のアルコールでアルコキシ化し、溶媒に溶解させていたが、反応工程が長くなる事、アルコキシ化反応は酸性で行うため縮合が進み分子量の制御が難かしくなる等の欠点があった。

【0006】またメラミン樹脂は高い硬度、高い耐熱性、高い光沢により、化粧材として主として家具の水平化粧面に従来より使われている。近年意匠性を増すため、硬化後の化粧板を後加工する、いわゆるポストフォーム加工が盛んに実施されている。また、メラミン化粧板は高い硬度をもつ反面、経時的なクラックの発生が見られる事があり、その改良が行われている。

【0007】そのためメラミン樹脂に各種共縮合性の化合物で変性したり、各種添加剤を加えることにより、ポストフォーム加工性を向上、また耐クラックの改良等が行なわれているが、樹脂への組み込みが不良であると耐水性の低下、光沢の低下となり、かならずしも満足のいくものでなかった。

【0008】またメラミンは尿素と組み合わせて木質系接着剤としてよく用いられている。同種用途に用いられているフェノール系接着剤と比較して硬化速度が早いことによる生産性にすぐれるが、メラミン系接着剤自体が親油性に乏しいため近年とみに用いられるヤニ分の多い針葉樹に対して不利があった。

【0009】

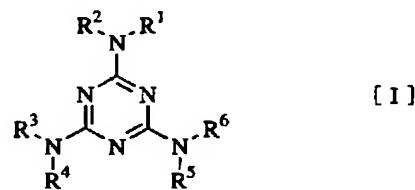
【発明が解決しようとする課題】本発明はメラミン樹脂の耐熱性、高い硬度、すぐれた光沢などの特性を損なうことなく、強靭性、曲げ加工性、親油性を与えるアミノ樹脂組成物を提供するものである。

【0010】

【課題を解決するための手段】本発明は、式【I】

【0011】

【化3】



【0012】【式中、置換基、R¹、R²、R³、R⁴、R⁵およびR⁶は各々互いに独立した置換基を表し、それら置換基の1ないし5個は、C₁₋₂₀のアルキル基、C₂₋₂₀のアルケニル基（該アルキル基またはアルケニル基は、その構造中に任意に脂環式構造またはフェニル基を有していても良く、また2個の該アルキル基またはアルケニル基が同一窒素原子上にある場合はその窒素原子と一緒にになって3～8員環を形成しても良い）また

はフェニル基を表し、そして残りの置換基は水素原子を表す。】により表されるトリアジン誘導体の1種または2種以上の混合物にホルムアルデヒドを反応させることにより製造されるアミノ樹脂組成物に関する。

【0013】更に本発明は、式【I】により表されるトリアジン誘導体の1種または2種以上の混合物およびメラミン又は尿素からなる混合物にホルムアルデヒドを反応させることにより製造されるアミノ樹脂組成物に関する。

10 【0014】そして、更に本発明は、式【I】により表されるトリアジン誘導体の1種または2種以上の混合物にホルムアルデヒドを反応させることにより製造されるアミノ樹脂組成物およびメラミン-ホルムアルデヒド樹脂、メラミン・尿素-ホルムアルデヒド樹脂又は尿素ホルムアルデヒド樹脂を含有するアミノ樹脂組成物に関する。以下、本発明を詳細に説明する。

【0015】

【発明の実施形態】本発明のアミノ樹脂組成物の製造のために使用される置換トリアジン誘導体は、下記の公知の合成方法と同様な方法で容易に得られる：例えばジャーナル・オブ・アメリカン・ケミカル・ソサエティ（J. Am. Chem. Soc.）73巻、2984頁、1951年に記載の2-クロロ-1, 3, 5-トリアジン誘導体とアルキルアミンを反応させて得る方法；

20 ヘミシェ・ペリヒテ（Chem. Ber.）18巻、2755頁、1885年に記載の2, 4, 6-トリメチルチオ-1, 3, 5-トリアジン誘導体とアルキルアミンを反応させて得る方法；米国特許第2, 228, 161号（1941年）に記載の2, 4, 6-トリアミノ-1, 3, 5-トリアジンとアルキルアミン塩酸塩を反応させて得る方法；ドイツ特許第889, 593号（1953年）に記載のシアノビペリジンとシアノグアニジンを反応させて2-ビペリジノ-4, 6-ジアミノ-1, 3, 5-トリアジンを得る方法；特開平3-215564号に記載の塩化シアヌルと相当するアルキルアミン類とを反応させて得る方法等が挙げられる。

30 【0016】外の方法としては、本発明の出願人により特許出願された下記の方法である：

(a)特開平8-27128号に記載の1, 3, 5-トリアジン誘導体と周期律表第VII族或いは第VIII族の存在下、アルコールと反応させる方法；

40 (b)特開平8-193071号に記載の1, 3, 5-トリアジン誘導体と周期律表第VII族或いは第VIII族の存在下でアルデヒドと反応させる方法；及び

(c)特開平8-27125号に記載の1, 3, 5-トリアジン誘導体と周期律表第VII族或いは第VIII族の存在下でオレフィンと反応させる方法等が挙げられる。

50 【0017】本発明のアミノ樹脂組成物の製造に使用さ

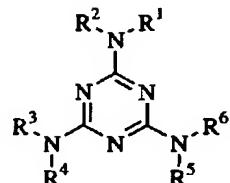
れるトリアジン誘導体は、上記のいずれの方法により製造される得るトリアジン誘導体であってもよい。しかし、電子材料分野に使用する場合は、置換トリアジン中に残留する加水分解性塩素の微小のもの乃至はないものが好ましい。従って、上記の文献(a)のメラミンに特定の触媒存在下にてアルコール類を反応させる方法、(b)のメラミンに水素雰囲気中で特定の触媒存在下にてアルデヒド類を反応させる方法、又は(c)のメラミンに水素/一酸化炭素雰囲気中で特定の触媒存在下にてオレフィン類を反応させる方法により得られるトリアジン誘導体が好ましい。

【0018】C₁₋₁₂のアルキル基、C₂₋₁₀のアルケニル基（該アルキル基またはアルケニル基は、その構造中に任意に脂環式構造またはフェニル基を有していても良く、また2個の該アルキル基またはアルケニル基が同一窒素原子上にある場合はその窒素原子と一緒にになって3~8員の含窒素複素環を形成しても良い）またはフェニル基で表される場合の置換基、R¹、R²、R³、R⁴、R⁵およびR⁶の具体例を下記する：メチル基、エチル基、n-プロピル基、iso-プロピル基、n-ブチル基、iso-ブチル基、sec-ブチル基、tert-ブチル基、シクロプロピルメチル基、シクロブチル基、n-ベンチル基、iso-ベンチル基、sec-ベンチル基、tert-ベンチル基、シクロベンチル基、n-ヘキシル基、シクロヘキシル基、シメ

Me : メチル基	E t : エチル基
P r : ノルマルプロピル基	ME : イソプロピル基
B u : ノルマルブチル基	I B : イソブチル基
M P : セカンダリープチル基	T B : ターシャリープチル基
P E : ノルマルベンチル基	H E : ノルマルヘキシル基
E H : 2-エチルヘキシル基	O c : ノルマルオクチル基
D e : ノルマルデシル基	D D : ノルマルドデシル基
H D : ノルマルヘキサデシル基	O D : ノルマルオクタデシル基
A L : アリル基	S T : スチリル基
C y : シクロヘキシル基	C M : シクロヘキシルメチル基
P h : フェニル基	B z : ベンジル基。

【0021】

【化4】



[I]

【0022】

【表1】

表1

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
Me	H	H	H	H	H

* クロヘキシルメチル基、4-メチルシクロヘキシルメチル基、n-オクチル基、2-エチルヘキシル基、n-ノニル基、n-デシル基、n-ドデシル基、n-ヘキサデシル基、n-オクタデシル基、ベンジル基、1-フェネチル基、2-フェネチル基、1-フェニルプロピル基、3-フェニルプロピル基、ビニル基、アリル基、メタリル基、クロチル基、2-ベンチニル基、3-ヘキセニル基、スチリル基及びフェニル基；そして、同一窒素原子上の2個の置換基がその窒素原子と一緒にになって複素環を形成した場合の環構造としてはアジリジン環、アゼチジン環、ビロリジン環及びビペリジン環等が挙げられる。なお、これら置換基の例示により、本発明のアミノ樹脂組成物の製造のために使用されるトリアジン誘導体の範囲が限定されるものではない。

10 【0019】本発明においては、上述の式[1]により表されるトリアジン誘導体の1種または2種以上を組合せて、ホルムアルデヒドとの反応、或いはメラミンまたは尿素とホルムアルデヒドとの反応に用いることが可能である。下記の表1に、本発明で用いられるトリアジン誘導体の主なものを、その置換基の組合せによって、さらに具体的に示す。

20 【0020】尚、表中の置換基の略号は、以下の意味を示す：

Me : メチル基	Me : メチル基	H : ハイドロキシル基	H : ハイドロキシル基	H : ハイドロキシル基	H : ハイドロキシル基
P r : ノルマルプロピル基	ME : イソプロピル基	I B : イソブチル基			
B u : ノルマルブチル基		T B : ターシャリープチル基			
M P : セカンダリープチル基		H E : ノルマルヘキシル基			
P E : ノルマルベンチル基		O c : ノルマルオクチル基			
E H : 2-エチルヘキシル基		D D : ノルマルドデシル基			
D e : ノルマルデシル基		H D : ノルマルヘキサデシル基			
H D : ノルマルヘキサデシル基		O D : ノルマルオクタデシル基			
A L : アリル基		S T : スチリル基			
C y : シクロヘキシル基		C M : シクロヘキシルメチル基			
P h : フェニル基		B z : ベンジル基。			

Me	Me	H	H	H	H
Me	H	Me	H	H	H
Me	Me	Me	H	H	H
Me	H	Me	H	Me	H
40 Me	Me	Me	Me	H	H
Me	Me	Me	H	Me	H
Me	Me	Me	Me	Me	H
E t	H	H	H	H	H
E t	E t	H	H	H	H
E t	H	E t	H	H	H
E t	E t	E t	H	E t	H
E t	H	E t	H	E t	H
E t	E t	E t	E t	H	H
E t	E t	E t	H	E t	H
50 E t	E t	E t	E t	E t	H

7

P r	H	H	H	H	H
P r	P r	H	H	H	H
P r	H	P r	H	H	H
P r	P r	P r	H	H	H
P r	H	P r	H	P r	H
P r	P r	P r	H	P r	H

【0023】

【表2】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
P r	P r	P r	P r	P r	H
ME	H	H	H	H	H
ME	ME	H	H	H	H
ME	H	ME	H	H	H
ME	ME	ME	H	H	H
ME	ME	ME	H	ME	H
ME	ME	ME	ME	ME	H
Bu	H	H	H	H	H
Bu	Bu	H	H	H	H
Bu	H	Bu	H	H	H
Bu	Bu	Bu	H	H	H
Bu	H	Bu	H	Bu	H
Bu	Bu	Bu	Bu	H	H
Bu	Bu	Bu	Bu	Bu	H
I B	H	H	H	H	H
I B	I B	H	H	H	H
I B	H	I B	H	H	H
I B	I B	I B	H	H	H
I B	H	I B	H	I B	H

【0024】

【表3】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
I B	I B	I B	I B	H	H
I B	I B	I B	H	I B	H
I B	I B	I B	I B	I B	H
MP	H	H	H	H	H
MP	MP	H	H	H	H
MP	H	MP	H	H	H
MP	MP	MP	H	H	H
MP	H	MP	H	MP	H

8

MP	MP	MP	MP	H	H
MP	MP	MP	H	MP	H
MP	MP	MP	MP	MP	H
T B	H	H	H	H	H
T B	T B	H	H	H	H
T B	H	T B	H	H	H
T B	T B	T B	H	H	H
T B	H	T B	H	T B	H
T B	T B	T B	T B	H	H
T B	T B	T B	T B	T B	H
PE	H	H	H	H	H
PE	PE	H	H	H	H
PE	H	PE	H	H	H

【0025】

【表4】

表1続き

20	R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
	PE	PE	PE	H	H	H
	PE	H	PE	H	PE	H
	PE	PE	PE	PE	H	H
	PE	PE	PE	H	PE	H
	PE	PE	PE	PE	PE	H
	HE	H	H	H	H	H
	HE	HE	H	H	H	H
	HE	H	HE	H	H	H
30	HE	HE	HE	H	H	H
	HE	H	HE	H	HE	H
	HE	HE	HE	HE	H	H
	HE	HE	HE	H	HE	H
	HE	HE	HE	HE	HE	H
	EH	H	H	H	H	H
	EH	EH	H	H	H	H
	EH	H	EH	H	H	H
	EH	EH	EH	H	H	H
	EH	H	EH	H	EH	H
40	EH	EH	EH	EH	H	H
	EH	EH	EH	H	EH	H
	EH	EH	EH	EH	EH	H
	O c	H	H	H	H	H

【0026】

【表5】

表1続き

50	R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
----	----------------	----------------	----------------	----------------	----------------	----------------

9	Oc	Oc	H	H	H	H
Oc	H	Oc	H	H	H	H
Oc	Oc	Oc	H	H	H	H
Oc	H	Oc	H	Oc	H	H
Oc	Oc	Oc	Oc	H	H	H
Oc	Oc	Oc	H	Oc	H	H
Oc	Oc	Oc	Oc	Oc	H	H
De	H	H	H	H	H	H
De	De	H	H	H	H	H
De	H	De	H	H	H	H
De	De	De	H	H	H	H
De	H	De	H	De	H	H
De	De	De	De	H	H	H
De	De	De	H	De	H	H
De	De	De	De	De	H	H
DD	H	H	H	H	H	H
DD	DD	H	H	H	H	H
DD	H	DD	H	H	H	H
DD	DD	DD	H	H	H	H
DD	H	DD	H	DD	H	H
DD	DD	DD	DD	H	H	H
DD	DD	DD	H	DD	H	H

【0027】

【表6】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
DD	DD	DD	DD	DD	H
HD	H	H	H	H	H
HD	HD	H	H	H	H
HD	H	HD	H	H	H
HD	HD	HD	H	H	H
HD	H	HD	H	HD	H
HD	HD	HD	HD	H	H
HD	HD	HD	H	HD	H
HD	HD	HD	HD	HD	H
OD	H	H	H	H	H
OD	OD	H	H	H	H
OD	H	OD	H	H	H
OD	OD	OD	H	H	H
OD	OD	OD	H	OD	H
OD	OD	OD	OD	H	H
OD	OD	OD	OD	OD	H
AL	H	H	H	H	H
AL	AL	H	H	H	H
AL	H	AL	H	H	H
AL	AL	AL	H	H	H

9	AL	H	AL	H	AL	H
【0028】						
【表7】						
表1続き						

10	R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
AL	AL	AL	AL	AL	H	H
AL	AL	AL	AL	H	AL	H
AL	AL	AL	AL	AL	AL	H
ST	H	H	H	H	H	H
ST	ST	H	H	H	H	H
ST	H	ST	H	H	H	H
ST	ST	ST	H	H	H	H
ST	H	ST	H	ST	H	H
ST	ST	ST	ST	H	ST	H
ST	ST	ST	ST	ST	ST	H
20	Cy	H	H	H	H	H
Cy	H	Cy	H	H	H	H
Cy	H	Cy	H	Cy	H	H
CM	H	H	H	H	H	H
CM	H	CM	H	H	H	H
CM	H	CM	H	CM	H	H
Ph	H	H	H	H	H	H
Ph	H	Ph	H	H	H	H
Ph	H	Ph	H	Ph	H	H
Bz	H	H	H	H	H	H
30	Bz	H	Bz	H	H	H

【0029】
【表8】
表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
Bz	H	Bz	H	Bz	H
DD	Me	H	H	H	H
40	DD	H	Me	H	H
DD	Me	Me	H	H	H
DD	H	Me	H	Me	H
DD	Me	Me	Me	H	H
DD	Me	Me	H	Me	H
DD	Me	Me	Me	Me	H
DD	Me	Me	Me	Me	H
DD	E _t	H	H	H	H
DD	H	E _t	H	H	H
DD	E _t	E _t	H	E _t	H
50	DD	E _t	E _t	H	H

11

DD	E t	E t	H	E t	H
DD	E t	E t	E t	E t	H
DD	P r	H	H	H	H
DD	H	P r	H	H	H
DD	P r	P r	H	H	H
DD	H	P r	H	P r	H
DD	P r	P r	H	P r	H
DD	P r	P r	P r	P r	H
DD	ME	H	H	H	H

【0030】

【表9】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
----------------	----------------	----------------	----------------	----------------	----------------

DD	H	ME	H	H	H
DD	ME	ME	H	H	H
DD	H	ME	H	ME	H
DD	ME	ME	ME	H	H
DD	ME	ME	H	ME	H
DD	ME	ME	ME	ME	H
DD	B u	H	H	H	H
DD	H	B u	H	H	H
DD	B u	B u	H	H	H
DD	H	B u	H	B u	H
DD	B u	B u	B u	H	H
DD	B u	B u	H	B u	H
DD	B u	B u	B u	B u	H
DD	I B	H	H	H	H
DD	H	I B	H	H	H
DD	I B	I B	H	H	H
DD	H	I B	H	I B	H
DD	I B	I B	I B	H	H
DD	I B	I B	H	I B	H
DD	I B	I B	I B	I B	H
DD	MP	H	H	H	H
DD	H	MP	H	H	H

【0031】

【表10】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
----------------	----------------	----------------	----------------	----------------	----------------

DD	MP	MP	H	H	H
DD	H	MP	H	MP	H
DD	MP	MP	MP	H	H
DD	MP	MP	H	MP	H
DD	MP	MP	MP	MP	H

12

DD	T B	H	H	H	H
DD	H	T B	H	H	H
DD	T B	T B	H	H	H
DD	H	T B	H	T B	H
DD	T B	T B	T B	H	H
DD	T B	T B	H	T B	H
DD	T B	T B	T B	T B	H
DD	P E	H	H	H	H
DD	H	P E	H	H	H

10

DD	P E	P E	H	H	H
DD	H	P E	H	P E	H
DD	P E	P E	P E	H	H
DD	P E	P E	H	P E	H
DD	P E	P E	P E	P E	H
DD	H E	H	H	H	H
DD	H	H E	H	H	H
DD	H E	H E	H	H	H

20

【表11】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
DD	H	HE	H	HE	H
DD	HE	HE	HE	H	H
DD	HE	HE	H	HE	H
DD	HE	HE	HE	HE	H
DD	EH	H	H	H	H
30	DD	H	EH	H	H
DD	EH	EH	H	H	H
DD	H	EH	H	EH	H
DD	EH	EH	EH	H	H
DD	EH	EH	H	EH	H
DD	EH	EH	EH	EH	H
DD	O c	H	H	H	H
DD	H	O c	H	H	H
DD	O c	O c	H	H	H
DD	H	O c	H	O c	H
40	DD	O c	O c	O c	H
DD	O c	O c	H	O c	H
DD	O c	O c	O c	O c	H
DD	De	H	H	H	H
DD	H	De	H	H	H
DD	De	De	H	H	H
DD	H	De	H	De	H

50

【表12】

表1続き

15

HD	H	I B	H	I B	H
HD	I B	I B	I B	H	H
HD	I B	I B	H	I B	H
HD	I B	I B	I B	I B	H
HD	MP	H	H	H	H
HD	H	MP	H	H	H
HD	MP	MP	H	H	H
HD	H	MP	H	MP	H
HD	MP	MP	MP	H	H
HD	MP	MP	H	MP	H
HD	MP	MP	MP	MP	H
HD	T B	H	H	H	H

【0037】

【表16】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
HD	H	T B	H	H	H
HD	T B	T B	H	H	H
HD	H	T B	H	T B	H
HD	T B	T B	T B	H	H
HD	T B	T B	H	T B	H
HD	T B	T B	T B	T B	H
HD	P E	H	H	H	H
HD	H	P E	H	H	H
HD	P E	P E	H	H	H
HD	H	P E	H	P E	H
HD	P E	P E	P E	H	H
HD	P E	P E	H	P E	H
HD	P E	P E	P E	P E	H
HD	H E	H	H	H	H
HD	H	H E	H	H	H
HD	H E	H	H	H	H
HD	H	H E	H	H E	H
HD	H E	H E	H E	H	H
HD	H E	H E	H	H E	H
HD	H E	H E	H E	H E	H
HD	E H	H	H	H	H
HD	H	E H	H	H	H

【0038】

【表17】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
HD	E H	E H	H	H	H
HD	H	E H	H	E H	H

16

HD	E H	E H	E H	H	H
HD	E H	E H	E H	E H	H
HD	E H	E H	E H	E H	H
HD	O c	H	H	H	H
HD	H	O c	H	H	H
HD	O c	O c	H	H	H
HD	H	O c	H	O c	H
HD	O c	O c	O c	H	H
10	HD	O c	O c	O c	H
HD	D e	H	H	H	H
HD	H	D e	H	H	H
HD	D e	D e	H	H	H
HD	H	D e	H	D e	H
HD	D e	D e	H	D e	H
HD	D e	D e	H	D e	H
HD	D e	D e	D e	D e	H
HD	D D	H	H	H	H
HD	H	D D	H	H	H
20	HD	D D	D D	H	H

【0039】

【表18】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
HD	H	D D	H	D D	H
HD	D D	D D	D D	H	H
30	HD	D D	D D	H	D D
HD	D D	D D	D D	D D	H
HD	O D	H	H	H	H
HD	H	O D	H	H	H
HD	O D	O D	H	H	H
HD	H	O D	H	O D	H
HD	O D	O D	O D	H	H
HD	O D	O D	H	O D	H
HD	O D	O D	O D	O D	H
HD	A L	H	H	H	H
40	HD	H	A L	H	H
HD	A L	A L	H	H	H
HD	H	A L	H	A L	H
HD	A L	A L	A L	H	H
HD	A L	A L	H	A L	H
HD	A L	A L	A L	A L	H
HD	S T	H	H	H	H
HD	H	S T	H	H	H
HD	S T	S T	H	H	H
50	H	S T	H	S T	H

【0040】

【表19】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
HD	ST	ST	ST	H	H
HD	ST	ST	H	ST	H
HD	ST	ST	ST	ST	H
HD	H	Cy	H	H	H
HD	H	Cy	H	Cy	H
HD	H	CM	H	H	H
HD	H	CM	H	CM	H
HD	H	Ph	H	H	H
HD	H	Ph	H	Ph	H
HD	H	Bz	H	H	H
HD	H	Bz	H	Bz	H
OD	Me	H	H	H	H
OD	H	Me	H	H	H
OD	Me	Me	H	H	H
OD	H	Me	H	Me	H
OD	Me	Me	Me	H	H
OD	Me	Me	H	Me	H
OD	Me	Me	Me	Me	H
OD	E t	H	H	H	H
OD	H	E t	H	H	H
OD	E t	E t	H	H	H
OD	H	E t	H	E t	H

【0041】

【表20】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
OD	E t	E t	E t	H	H
OD	E t	E t	H	E t	H
OD	E t	E t	E t	E t	H
OD	P r	H	H	H	H
OD	H	P r	H	H	H
OD	P r	P r	H	H	H
OD	H	P r	H	P r	H
OD	P r	P r	H	P r	H
OD	P r	P r	P r	P r	H
OD	ME	H	H	H	H
OD	H	ME	H	H	H
OD	ME	ME	H	H	H
OD	H	ME	H	ME	H
OD	ME	ME	ME	H	H
OD	ME	ME	H	ME	H

OD	ME	ME	ME	ME	H
OD	B u	H	H	H	H
OD	H	B u	H	H	H
OD	B u	B u	H	H	H
OD	H	B u	H	B u	H
OD	B u	B u	B u	H	H
OD	B u	B u	H	B u	H

10 【0042】
【表21】
表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
OD	B u	B u	B u	B u	H
OD	I B	H	H	H	H
OD	H	I B	H	H	H
OD	I B	I B	H	H	H
OD	H	I B	H	I B	H
20	OD	I B	I B	I B	H
OD	I B	I B	H	I B	H
OD	I B	I B	I B	I B	H
OD	MP	H	H	H	H
OD	H	MP	H	H	H
OD	MP	MP	H	H	H
OD	H	MP	H	MP	H
OD	MP	MP	MP	H	H
OD	MP	MP	MP	MP	H
OD	MP	MP	MP	MP	H
30	OD	T B	H	H	H
OD	H	T B	H	H	H
OD	T B	T B	H	H	H
OD	H	T B	H	T B	H
OD	T B	T B	T B	H	H
OD	T B	T B	H	T B	H
OD	T B	T B	T B	T B	H

30 【0043】
【表22】
40 表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
PE	PE	H	H	H	H
OD	H	PE	H	H	H
OD	PE	PE	H	H	H
OD	H	PE	H	PE	H
OD	PE	PE	PE	H	H
OD	PE	PE	H	PE	H
OD	PE	PE	PE	PE	H
50	OD	PE	PE	PE	H

19					
OD	HE	H	H	H	H
OD	H	HE	H	H	H
OD	HE	HE	H	H	H
OD	H	HE	H	HE	H
OD	HE	HE	HE	H	H
OD	HE	HE	H	HE	H
OD	HE	HE	HE	HE	H
OD	EH	H	H	H	H
OD	H	EH	H	H	H
OD	EH	EH	H	H	H
OD	H	EH	H	EH	H
OD	EH	EH	EH	H	H
OD	EH	EH	H	EH	H
OD	EH	EH	EH	EH	H
OD	Oc	H	H	H	H

【0044】

【表23】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
OD	H	Oc	H	H	H
OD	Oc	Oc	H	H	H
OD	H	Oc	H	Oc	H
OD	Oc	Oc	Oc	H	H
OD	Oc	Oc	H	Oc	H
OD	Oc	Oc	Oc	Oc	H
OD	De	H	H	H	H
OD	H	De	H	H	H
OD	De	De	H	H	H
OD	H	De	H	De	H
OD	De	De	De	H	H
OD	De	De	H	De	H
OD	De	De	De	De	H
OD	DD	H	H	H	H
OD	H	DD	H	H	H
OD	DD	DD	H	H	H
OD	H	DD	H	DD	H
OD	DD	DD	DD	H	H
OD	DD	DD	H	DD	H
OD	DD	DD	DD	DD	H
OD	HD	H	H	H	H
OD	H	HD	H	H	H

【0045】

【表24】

表1続き

R¹ R² R³ R⁴ R⁵ R⁶

20						
OD	HD	HD	H	H	H	
OD	H	HD	H	HD	H	
OD	HD	HD	HD	H	H	
OD	HD	HD	H	HD	H	
OD	HD	HD	HD	HD	H	
OD	AL	H	H	H	H	
OD	H	AL	H	H	H	
OD	AL	AL	H	H	H	
10	OD	H	AL	H	AL	H
OD	AL	AL	AL	H	H	
OD	AL	AL	H	AL	H	
OD	AL	AL	AL	AL	H	
OD	ST	H	H	H	H	
OD	H	ST	H	H	H	
OD	ST	ST	H	H	H	
OD	H	ST	H	ST	H	
OD	ST	ST	ST	H	H	
OD	ST	ST	H	ST	H	
20	OD	ST	ST	ST	H	
OD	H	Cy	H	H	H	
OD	H	Cy	H	Cy	H	
OD	H	CM	H	H	H	

【0046】

【表25】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶	
30	OD	H	CM	H	CM	H
OD	H	Ph	H	H	H	
OD	H	Ph	H	Ph	H	
OD	H	Bz	H	H	H	
OD	H	Bz	H	Bz	H	
Me	H	EH	H	H	H	
Me	Me	EH	H	H	H	
Me	H	EH	H	Me	H	
Me	Me	EH	H	Me	H	
40	Me	Me	Me	Me	EH	H
Me	H	Cy	H	H	H	
Me	Me	Cy	H	H	H	
Me	H	Cy	H	Me	H	
Me	Me	Cy	H	Me	H	
Me	Me	Me	Me	Cy	H	
Me	H	Bz	H	H	H	
Me	Me	Bz	H	H	H	
Me	H	Bz	H	Me	H	
Me	Me	Bz	H	Me	H	
50	Me	Me	Me	Me	Bz	H

21

E t	H	E H	H	H	H
E t	E t	E H	H	H	H

【0047】

【表26】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
E t	H	E H	H	E t	H
E t	E t	E H	H	E t	H
E t	E t	E t	E t	E H	H
E t	H	C y	H	H	H
E t	E t	C y	H	H	H
E t	H	C y	H	E t	H
E t	E t	C y	H	E t	H
E t	E t	E t	E t	C y	H
E t	H	B z	H	H	H
E t	E t	B z	H	H	H
E t	H	B z	H	E t	H
E t	E t	B z	H	E t	H
E t	E t	E t	E t	B z	H
ME	H	E H	H	H	H
ME	H	C y	H	H	H
ME	H	C y	H	ME	H
ME	H	B z	H	H	H
ME	H	B z	H	ME	H
Bu	H	E H	H	H	H
Bu	B u	E H	H	H	H
Bu	H	E H	H	B u	H
Bu	B u	E H	H	B u	H

【0048】

【表27】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
Bu	B u	B u	B u	E H	H
Bu	H	C y	H	H	H
Bu	B u	C y	H	H	H
Bu	H	C y	H	B u	H
Bu	B u	C y	H	B u	H
Bu	B u	B u	B u	C y	H
Bu	H	B z	H	H	H
Bu	B u	B z	H	H	H
Bu	H	B z	H	B u	H
Bu	B u	B z	H	B u	H
Bu	B u	B u	B u	B z	H
AL	H	E H	H	H	H

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AL	AL	E H	H	H	H
AL	H	E H	H	AL	H
AL	AL	E H	H	AL	H
AL	AL	AL	AL	E H	H
AL	H	C y	H	H	H
AL	AL	C y	H	H	H
AL	H	C y	H	AL	H
AL	AL	C y	H	AL	H
AL	H	B z	H	H	H
10	AL	AL	B z	H	H

【0049】

【表28】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
AL	H	B z	H	AL	H
AL	AL	B z	H	AL	H
AL	AL	AL	AL	B z	H
-(CH ₂) ₄ -		H	H	H	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -		H	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -		B u	H
-(CH ₂) ₅ -		H	H	H	H
-(CH ₂) ₅ -		H	H	M e	H
-(CH ₂) ₅ -		H	H	E t	H
-(CH ₂) ₅ -		H	H	P r	H
-(CH ₂) ₅ -		H	H	B u	H
-(CH ₂) ₅ -		-(CH ₂) ₅ -		H	H
-(CH ₂) ₅ -		-(CH ₂) ₅ -		B u	H
-(CH ₂) ₄ -		E H	H	H	H
-(CH ₂) ₄ -		E H	H	B u	H
-(CH ₂) ₄ -		E H	H	M P	H
-(CH ₂) ₄ -		E H	H	P E	H
-(CH ₂) ₄ -		E H	H	H E	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -		E H	H
-(CH ₂) ₄ -		C y	H	H	H
-(CH ₂) ₄ -		C y	H	I B	H
-(CH ₂) ₄ -		C y	H	E H	H
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【0050】

【表29】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
-(CH ₂) ₄ -	Cy	H	Oc	H	
-(CH ₂) ₄ -	Cy	H	OD	H	
-(CH ₂) ₄ -		-(CH ₂) ₄ -	Cy	H	
-(CH ₂) ₄ -	Bz	H	H	H	
-(CH ₂) ₄ -	Bz	H	Me	H	
-(CH ₂) ₄ -	Bz	H	Et	H	
-(CH ₂) ₄ -	Bz	H	Pr	H	
-(CH ₂) ₄ -	Bz	H	Bu	H	
-(CH ₂) ₄ -		-(CH ₂) ₄ -	Bz	H	
-(CH ₂) ₅ -	EH	H	H	H	
-(CH ₂) ₅ -	EH	H	Me	H	
-(CH ₂) ₅ -	EH	H	Et	H	
-(CH ₂) ₅ -	EH	H	Pr	H	
-(CH ₂) ₅ -	EH	H	EH	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	EH	H	
-(CH ₂) ₅ -	Cy	H	H	H	
-(CH ₂) ₅ -	Cy	Bu	H	H	
-(CH ₂) ₅ -	Cy	HE	H	H	
-(CH ₂) ₅ -	Cy	H	De	H	
-(CH ₂) ₅ -	Cy	H	HD	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	H	H	
-(CH ₂) ₅ -	Bz	H	H	H	

【0051】

【表30】

表1続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
-(CH ₂) ₅ -	Bz	De	H	H	
-(CH ₂) ₅ -	Bz	HD	H	H	
-(CH ₂) ₅ -	Bz	H	De	H	
-(CH ₂) ₅ -	Bz	H	HD	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	Bz	H	

【0052】上記の式【I】の置換基の組み合わせの中、好ましいものは、置換基、R¹、R²、R³、R⁴、R⁵およびR⁶のうち、R¹、R³およびR⁵の1ないし3個がC₁₋₁₀のアルキル基を表し、残りの置換基が水素原子を表す群である。

【0053】上記の式【I】の置換基の組み合わせの中、更に好ましいものは、置換基、R¹、R²、R³、R⁴、R⁵およびR⁶のうち、R¹、R³およびR⁵の1ないし3個がメチル基、エチル基、n-ブロビル基、イソブロビル基、n-ブチル基、シクロヘキシル基又は2-エチルヘキシル基を表し、残りの置換基が水素原子

を表す群である。

【0054】アミノ樹脂組成物を製造するための、式【I】により表されるトリアジン誘導体の1種又は2種以上の混合物とホルムアルデヒドとの反応において、トリアジン誘導体の1種又は2種以上の混合物1モルに対するホルムアルデヒドのモル数はトリアジン誘導体1モルに対し1モルからトリアジン誘導体のアミノ基の未置換水素原子と同数のモル数が好ましい。ホルムアルデヒドのモル数が1より少ないと樹脂組成物の熱硬化により生成した構造体が充分なる架橋構造を形成し得ず、アミノ樹脂組成物の熱硬化物の耐熱性、高い硬度、強靭性等の物性が満足できるものとはならない。他方トリアジン誘導体のアミノ基の未置換水素原子と同数のモル数よりも多いと、トリアジン誘導体と付加反応にあずからなかったホルムアルデヒドが残存する。そして、残存するホルマリンの除去が難かしく、そのため、アミノ樹脂組成物のホルマリン臭が著しく好ましくない。熱硬化の際のホルマリン臭も著しい。又過剰のホルムアルデヒドの使用は経済的に不利である。

【0055】本発明のトリアジン誘導体とホルムアルデヒドとの反応は通常水性媒体中で行われるが、本発明のトリアジン誘導体は通例、有機溶媒に可溶のため水にも可溶な有機溶媒を加えるとホルムアルデヒドとの反応が円滑に進む。有機溶媒としてはメタノール、エタノール、イソブロバノール、ブロバノールなどのアルコール類、ジオキサン、テトラヒドロフラン、1,2-ジメトキシエタン等のエーテル類、ジメチルホルムアミド、ジメチルスルホキシドなどの極性溶媒が好ましい。

【0056】トリアジン誘導体とホルムアルデヒドとの反応である樹脂化反応において、通常、反応温度は50～95°Cの範囲内である。しかし、この範囲より低い温度でも反応は進行することがあり、例えば有機溶媒を加えた場合は、50°Cより低温で樹脂化反応が進行する。

【0057】樹脂化反応時の反応液のpHは6～10、好適には7～9である。樹脂化反応による生成物の分子量を増大させるためにはpHは中性側、例えば7～7.5であり、分子量を低く抑える場合にはアルカリ側例えばpH 8～9で樹脂化反応を進行させる。

【0058】反応時間、即ち樹脂化反応開始から反応終了に至るまでの時間は、所望の分子量の樹脂組成物が得られる時点を反応終了時点とすることにより決められる。分子量は、反応中適宜サンプリングし、白濁温度法、水混和度法および/またはゲルバーミュエーションクロマト法による分子量測定により決定される。上述のように、本発明では、式【I】により表されるトリアジン誘導体の1種または2種以上の混合物とホルムアルデヒドとの反応において、トリアジン誘導体の1種または2種以上の混合物にメラミンまたは尿素を混合させて樹脂化することも行われる。

【0059】トリアジン誘導体の1種または2種以上の

混合物に混合させるメラミンまたは尿素のトリアジン誘導体に対する量の制限はないが一般的にいって含有させるメラミンまたは尿素の量が大きくなると架橋密度が高くなり、硬度は増すが、脆くなり強靭性が低下する。含有させるメラミンまたは尿素のモル量は、トリアジン誘導体1モルに対して0.05から20モル、好ましくは0.1モルから10モルである。

【0060】メラミンまたは尿素と混合した式【1】に表されるトリアジン誘導体の1種又は2種以上の混合物とホルムアルデヒドの反応における反応条件、即ち溶媒、反応温度、反応液のpH、反応温度等は、上述したトリアジン誘導体とホルムアルデヒドとの反応のそれらと同じである。この反応におけるホルムアルデヒドのモル数は、メラミン、尿素とトリアジン誘導体の合計モル数に対し等しいモル数からメラミン、尿素、トリアジン誘導体のアミノ基の未置換水素原子と同数のモル数が好ましい。等しいモル数より少ないと、架橋が充分に形成し得ず物性が劣る。他方メラミン、尿素、トリアジン誘導体のアミノ基の未置換水素原子と同数のモル数より多いと、アミノ基との付加反応又は縮合反応にあずからなかつたホルムアルデヒドが残存し、又この除去が難かしいため、組成物にホルマリン臭が著しく好ましくない。又過剰のホルムアルデヒドの使用は経済的に不利である。

【0061】又、本発明の式【1】により表されるトリアジン誘導体の1種又は2種以上の混合物とホルムアルデヒドとを反応させて得られるアミノ樹脂組成物とメラミン-ホルムアルデヒド樹脂、メラミン-尿素-ホルムアルデヒド樹脂又は尿素-ホルムアルデヒド樹脂との混合物を用いることも行われる。混合物とは本発明のトリアジン誘導体の1種又は2種以上の混合物とホルムアルデヒドとを反応させて得られるアミノ樹脂組成物の液状品又は固体状品と、公知の方法で得られた中から選ばれた少なくとも1種のメラミン-ホルムアルデヒド樹脂、メラミン-尿素-ホルムアルデヒド樹脂又は尿素-ホルムアルデヒド樹脂の液状品又はその固体状品を任意の割合で混合してなるものをいう。

【0062】公知の方法で得られるメラミン-ホルムアルデヒド樹脂の例示としては、メラミン1モル、ホルムアルデヒド1.6モルをpH8で温度95°C時間60分程度反応させて得た固体分濃度55%、分子量600程度の樹脂液である。又、メラミン-尿素-ホルムアルデヒド樹脂の例示としてはメラミン0.2モル、尿素1モル、ホルムアルデヒド1.8モルをpH7.9、温度90°C、時間120分程度反応させて得た固体分濃度55%、分子量700程度の樹脂液である。更に又、尿素樹脂の例示としては、尿素1モル、ホルムアルデヒド1.8モルをpH8.0、温度90°C、時間150分程度反応させて得た固体分濃度50%、分子量500程度の樹脂液である。

【0063】トリアジン誘導体-ホルムアルデヒド樹脂組成物と前記のメラミン-ホルムアルデヒド樹脂、メラミン-尿素-ホルムアルデヒド樹脂及び尿素-ホルムアルデヒド樹脂の混合割合は、広い範囲のものであるが、トリアジン誘導体-ホルムアルデヒド樹脂組成物の割合が高いと強靭性、曲げ加工性、親油性等に与える改良効果が大きく、一方割合が低いと改良効果が乏しくなる。

配合量は、用途、目的によって異なるが、トリアジン誘導体-ホルムアルデヒド樹脂組成物100重量部に対してメラミン-ホルムアルデヒド樹脂又はメラミン-尿素-ホルムアルデヒド樹脂又は尿素-ホルムアルデヒド樹脂は20~1000重量部、好適には、50~500重量部である。

【0064】上述のようにして製造されたアミノ樹脂組成物は木質材料接着剤、成形材、化粧材、塗料、等の分野に使用される。木質材料の接着剤として使用される場合は、硬化剤を混合し、木材の上に塗布あるいは木粉と混合し熱圧プレスによりアミノ樹脂組成物が熱硬化される。その際必要に応じ増粘剤、浸透コントロール剤等が添加される。

【0065】成形材として使用される場合は、液状のまま、あるいは乾燥粉末にした後、硬化剤を添加し、木粉、パルプ粉、無機粉末等と混合し熱圧成形により付形されると共にアミノ樹脂組成物が熱硬化される。化粧材として使用される場合は、硬化剤を添加したアミノ樹脂組成物樹脂液を紙等に含浸し乾燥され、フェノール樹脂含浸紙あるいは木質板と組み合わせ熱圧プレスによりアミノ樹脂組成物が熱硬化される。

【0066】塗料に使用される場合は、アミノ樹脂組成物はアルキッド樹脂、ポリエステル樹脂、アクリル樹脂等と組み合わせ硬化剤及びその他の添加剤を添加し塗料組成とし、加熱により組み合わせた樹脂のOH基と反応する架橋剤として使用される。その際、塗料の要求形態により、水溶性型、有機溶剤溶解型、固体型等の樹脂とされる。

【0067】

【実施例】以下に実施例及び比較例により本発明を更に詳細に説明するが、これらの実施例の記載は本発明の範囲の限定を意図するものではない。実施例に用いるトリアジン誘導体は以下の参考例の方法により合成した。

参考例1：2,4-ジアミノ-6-ノルマルプチルアミノ-1,3,5-トリアジンの合成

【0068】(a) 2,4-ジアミノ-6-クロル-1,3,5-トリアジンの合成

塩化シアヌール184.5g(1.0モル)をアセトニトリル800mLに室温にて溶解後、0°Cに冷却した溶液に、激しく攪拌しながら28%アンモニア水溶液303.7g(5.0モル)を反応温度を10°C以下を保つように、2時間で滴下した。滴下終了後、冷却を停止し室温で1時間攪拌した後に、徐々に加温して45°Cとし

て更に4時間反応させた。冷却後、生成物を口別し、さらに大量の水にて洗浄した。口過物を、真空下、50°Cで6時間乾燥することで、表記化合物を115g（収率79%）を得た。

【0069】(b) 2, 4-ジアミノ-6-ノルマルブチルアミノ-1, 3, 5-トリアジンの合成

(a)で合成した2, 4-ジアミノ-6-クロル-1, 3, 5-トリアジン14.5g(0.1モル)、水100mLおよびブチルアミン29.2g(0.4モル)の混合溶液を、攪拌しながら加温して、最終的に還流温度にて6時間反応させた。反応液を冷却後、生成物を口別し、さらに大量の水で充分に洗浄し、次にトルエンで洗浄した。口過物を、真空下、70°Cで6時間乾燥することで、表記の化合物を17.5g（収率96%）を得た。融点：167°C。

【0070】参考例2：2, 4, 6-トリス（ノルマルブチルアミノ）-1, 3, 5-トリアジンの合成

攪拌機、温度計を備えたオートクレーブに、メラミン12.6g(0.1モル)、1, 4-ジオキサン200g、n-ブチルアルデヒド72.0g(1.0モル)、5%Pd担持活性炭2.0gを仕込み、窒素ガス置換後、水素ガスを初期圧40kg/cm²として、激しく攪拌しながら反応温度180°Cにて6時間反応させた。反応終了後、室温までゆっくり冷却し、口別して触媒及び固体物を除去した後、溶剤を留去し、粘ちような液状の反応生成物の粗物を得た。これをシリカゲルカラムクロマトグラフィーにてアセトン/ヘキサン混合溶剤を溶離液として、混合比率を100/1~1/100に逐次濃度を変えつつ展開し、生成物を単離した後、溶剤を留去して、液状の表記化合物19.6gを得た。

【0071】参考例3：4, 6-ジアミノ-2-シクロヘキシルアミノ-1, 3, 5-トリアジンの合成

参考例1で合成した中間体の2, 4-ジアミノ-6-クロル-1, 3, 5-トリアジン14.5g(0.1モル)、水140mLおよびシクロヘキシルアミン29.2g(0.3モル)の混合溶液を、攪拌しながら加温して、還流温度にて1時間反応させた。さらに水酸化ナトリウム12gの水40mL溶液を1時間かけて滴下し、1時間熟成した。得られた反応混合物にトルエン200mLを加え、その後に室温に冷却した。得られた結晶を濾取し、トルエン100mL、続いて水100mLで順次洗浄後、減圧下に乾燥することで、表記化合物を17.9g（収率86%）を得た。融点：151°C。

【0072】参考例4：4, 6-ジアミノ-2-(2-エチルヘキシルアミノ)-1, 3, 5-トリアジンの合成

シクロヘキシルアミン29.2g(0.3モル)に換えて2-エチルヘキシルアミン12.9g(0.1モル)を用いる以外は、参考例3と同様に合成した。得量18.6g（収率78%）。融点：81°C。

【0073】参考例5：2, 4, 6-トリス（メチルアミノ）-1, 3, 5-トリアジンの合成

塩化シアヌール184.5g(1.0モル)をトルエン920gに室温で溶解後、0°Cに冷却した溶液に、激しく攪拌しながら40%のメチルアミン水溶液308g(4.0モル)滴下する。滴下終了後、攪拌しつつ2時間0°C保持した後、徐々に加温して30°Cとしてさらに1時間攪拌を行った。その溶液を更に40°Cに昇温し2時間攪拌し、70°Cまで昇温した。その溶液を70°Cに保持しつつジオキサンを46gと40%のメチルアミン水溶液464g(6.0モル)滴下し9時間攪拌を続けた。30°Cに冷却後40%水酸化ナトリウム水溶液を300g(3.0モル)を加え1時間攪拌し、更に5°Cに冷却し2時間攪拌した。生成物を濾別し水で洗浄した。濾過物を50°Cで6時間乾燥することで、表記化合物を93g（収率55%）を得た。融点133°C。

【0074】実施例1

参考例1の方法で合成した2, 4-ジアミノ-6-ノルマルブチルアミノ-1, 3, 5-トリアジン455g、40%ホルマリン300g、水184.3g、ジオキサン455gを攪拌機、温度計及び冷却管を備えた2L4つ口フラスコに仕込み5%KOHにてpHを8.0に調整した。その後、攪拌しながら、昇温し、温度を70°Cに保ち、2時間反応させ、その後室温まで冷却した。

【0075】40°Cで減圧濃縮し、黄色透明油状の樹脂組成物を540gを得た。ゲルバーミュエーションクロマト法(GPC法)で測定した分子量は580であった。

【0076】実施例2

参考例2に記載の方法で得た2, 4, 6-トリス（ノルマルブチルアミノ）-1, 3, 5-トリアジン264.6g、40%ホルマリン150g、水50g、ジオキサン150gを2L4つ口フラスコに仕込んだ。実施例1に準拠し、褐色油状の樹脂組成物300gを得た。GPC法で測定した分子量は520であった。

【0077】実施例3

参考例3に記載の方法で得た4, 6-ジアミノ-2-シクロヘキシルアミノ-1, 3, 5-トリアジン520g、40%ホルマリン300g、水184.3g、ジオキサン520gを2L4つ口フラスコに仕込み、実施例1に準拠し油状の樹脂組成物を589gを得た。GPC法による分子量は720であった。

【0078】実施例4

参考例4の方法で得た4, 6-ジアミノ-2-(2-エチルヘキシルアミノ)-1, 3, 5-トリアジン476g、40%ホルマリン240g、水147.5g、ジオキサン455gを2L4つ口フラスコに仕込み、実施例1に準拠しワックス状の樹脂組成物を得た。GPC法による分子量は670であった。

【0079】実施例5

メラミン63g、参考例1の方法で合成した2,4-ジアミノ-6-ノルマルブチルアミノ-1,3,5-トリアジン91g、40%ホルマリン120g、水50g、ジオキサン181gを攪拌機、温度計及び冷却管を備えた1L4つ口フラスコに仕込み、10%NaOH水溶液にてpHを9.2に調整した。その後、攪拌しながら、昇温し、温度を80°Cに保ち90分間反応させた。その後室温まで冷却し、透明液を得た。GPC法による分子量は850であった。

【0080】実施例6

参考例1の方法で合成した2,4-ジアミノ-6-ノルマルブチルアミノ-1,3,5-トリアジン455g、40%ホルマリン300g、水184.3g、ジオキサン455gを攪拌機、温度計及び冷却管を備えた2L4つ口フラスコに仕込み5%KOH水溶液にてpHを8.4に調整した。その後、攪拌しながら、昇温し、温度を70°Cに保ち2時間反応させた。

【0081】室温まで冷却し、冷却後5%KOHにてpHを9.0に調整し樹脂液を得た。この樹脂液500gとメラミン樹脂液(日産化学工業社製商品名サントップ・M700樹脂固形分55%)500gとを混合した。樹脂液は室温で1ヶ月保存して安定であった。

【0082】実施例7

参考例5の方法で合成した2,4,6-トリス(メチルアミノ)-1,3,5-トリアジン420g、40%ホルマリン300g、水360gを2L4つ口フラスコに仕込み、実施例1に準拠し反応させた。生成物のGPC法による分子量は420であった。

【0083】比較例1

メラミン1260g、40%ホルマリン1200g、水*

表2

	実施例						比較例
	1	2	3	4	5	6	
屈曲性 F(%)	48	65	52	60	40	41	9

【0088】評価例2

実施例7で得た樹脂液200gとメラミン樹脂液(日産化学工業株式会社製商品名サントップM700固形分55%)800gとを混合した樹脂液、及びメラミン樹脂液(日産化学工業株式会社製商品名サントップM700固形分55%)単独に、それぞれバラトルエンスルホン酸30%水溶液を、樹脂液に対し3%添加し含浸用の樹脂液とした。それぞれの樹脂液を80g/m²の化粧紙に含浸させ、105°Cで乾燥後、樹脂の付着量60%の含浸紙を得た。

【0089】得られたそれぞれの含浸紙を3枚積層し50°C、圧力10kg/cm²、10分間熱圧プレスし0.5mmの積層硬化シートを得た。それぞれの積層

*1400gを攪拌機、温度計及び冷却管を備えた5L4つ口フラスコに仕込み10%KOHにて、pHを8.4に調整した。その後、攪拌しながら、昇温し、温度を95°Cに保ち、60分反応させた。その後室温まで冷却し、10%KOH水溶液にてpHを9.0に調整し透明水性樹脂を得た。GPC法で測定した分子量は680であった。

【0084】評価例1

実施例1~4で得た樹脂液に少量のアセトンと硬化剤としてバラトルエンスルホン酸の30%アセトン溶液を樹脂液に対し3重量%加え低粘度液体とした。その液体を化学滤紙(東洋滤紙社製商品名5C)に含浸し、その後105°Cで乾燥させ、アセトンを除去し、滤紙の重さに対する樹脂の付着量を60%とした含浸紙を得た。実施例5、6及び比較例1についてはそれぞれの樹脂液を滤紙に含浸させ、105°Cで乾燥後、樹脂の付着量を60%とする含浸紙を得た。

【0085】得られたそれぞれの含浸紙を5枚積層し150°C、圧力10kg/cm²、10分間熱プレスし厚み0.8mmの積層硬化シートを得た。積層シートを長さ40mm、幅25mm、厚み0.8mmの短冊型のサンプルを切り出し、それを強度試験機(オリエンテック社製商品名テンションUCT-10T)に直立させ、屈曲速度5mm/分で破断するまで折り曲げた。

【0086】元の長さをL₀とし、破断した時の長さをL₁として屈曲性FをF=(L₀-L₁)/L₀×100/L₀として求めた。結果を表2に示す。

【0087】

【表31】

【0088】評価例2

実施例7で得た樹脂液200gとメラミン樹脂液(日産化学工業株式会社製商品名サントップM700固形分55%)800gとを混合した樹脂液、及びメラミン樹脂液(日産化学工業株式会社製商品名サントップM700固形分55%)単独に、それぞれバラトルエンスルホン酸30%水溶液を、樹脂液に対し3%添加し含浸用の樹脂液とした。それぞれの樹脂液を80g/m²の化粧紙に含浸させ、105°Cで乾燥後、樹脂の付着量60%の含浸紙を得た。

【0089】得られたそれぞれの含浸紙を3枚積層し50°C、圧力10kg/cm²、10分間熱圧プレスし0.5mmの積層硬化シートを得た。それぞれの積層

接着等の分野へも利用出来る。

【図面の簡単な説明】

【図1】 本発明の屈曲性試験において加重をかける前の状態を示す。

【図2】 本発明の屈曲性試験において加重をかけてい*

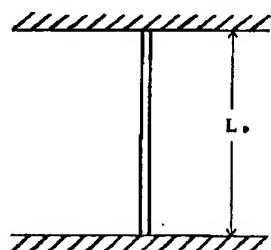
*る状態を示す。

【符号の説明】

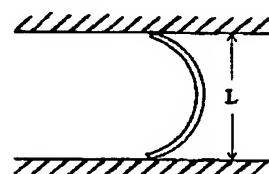
L₀ 加重をかける前の長さ。

L 加重をかけている時の長さ。

【図1】



【図2】



フロントページの続き

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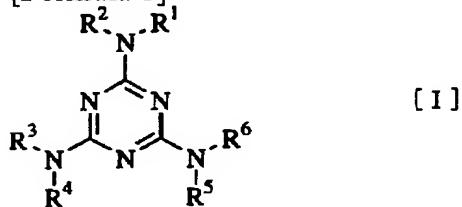
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CLAIMS

[Claim(s)]

[Claim 1] Formula [I]

[Formula 1]

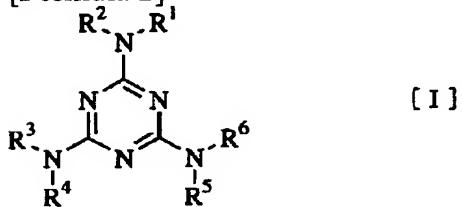


A substituent, R1, R2, R3, R4, R5, and R6 have been independent mutually respectively among [formula. 1 of these substituents or five pieces The alkyl group of C1-20, the alkenyl machine of C2-20 (this alkyl group or an alkenyl machine) You may have alicyclic structure or the phenyl group arbitrarily in the structure. moreover, when these two alkyl groups or an alkenyl machine is on the same nitrogen atom, or it may become together with the nitrogen atom and may form the nitrogen-containing heterocycle of 3 - 8 member, a phenyl group is expressed and the remaining substituent expresses a hydrogen atom] The amino resin constituent characterized by being manufactured by making formaldehyde react to one sort or two sorts or more of mixture of a triazine derivative which is alike and is expressed more.

[Claim 2] The amino resin constituent according to claim 1 manufactured by making the non-replaced hydrogen atom of the amino group of one sort or two sorts or more of mixture of this triazine derivative, and the formaldehyde of the mol of the same number react from one mol to one mol of one sort or two sorts or more of mixture of a triazine derivative expressed by the formula [I].

[Claim 3] Formula [I]

[Formula 2]



A substituent, R1, R2, R3, R4, R5, and R6 have been independent mutually respectively among [formula. 1 of these substituents or five pieces The alkyl group of C1-20, the alkenyl machine of C2-20 (this alkyl group or an alkenyl machine) You may have alicyclic structure or the phenyl group arbitrarily in the structure. moreover, when these two alkyl groups or an alkenyl machine is on the same nitrogen atom, or it may become together with the nitrogen atom and may form the nitrogen-containing heterocycle of 3 - 8 member, a phenyl group is expressed and the remaining substituent expresses a

hydrogen atom] The amino resin constituent characterized by being manufactured by making formaldehyde react to the mixture which consists of one sort or two sorts or more of the mixture and melamines, or ureas of a triazine derivative which are alike and are expressed more.

[Claim 4] The amino resin constituent containing an amino resin constituent according to claim 1 and a melamine formaldehyde resin, a melamine and a formaldehyde resin, or a formaldehyde resin.

[Claim 5] A substituent, R1, R2, R3, R4, and R5 And R6 Inside, R1, and R3 And R5 Amino resin constituent according to claim 1 to 4 with which 1 or three pieces express the alkyl group of C1-10, and the remaining substituent expresses a hydrogen atom.

[Claim 6] A substituent, R1, R2, R3, R4, and R5 And R6 Inside, R1, and R3 And R5 Amino resin constituent according to claim 5 with which 1 or three pieces express a methyl group, an ethyl group, n-propyl group, an isopropyl group, n-butyl, a cyclohexyl group, or a 2-ethylhexyl group, and the remaining substituent expresses a hydrogen atom.

[Claim 7] The panel containing an amino resin constituent according to claim 1 to 6, a laminate, a molding material, a paint, or adhesives.

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DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[The technical field to which invention belongs] this invention relates to the amino resin constituent excellent in tough nature, thermal resistance, lightfastness, resistance to contamination, etc. this invention constituent is useful in a molding material, makeup material, the curing agent for paints, woody system adhesives, etc.

[0002]

[Description of the Prior Art] Melamine resin is conventionally used for a molding material, makeup material, the curing agent for paints, woody system adhesives, etc. widely for the reasons nil why mold releasability with metal mold is good etc., at the time of that the outstanding thermal resistance, a high degree of hardness, high gloss, and a cure rate are early and fabrication.

[0003] On the other hand, since melamines were that it is six functionality and that a triazine ossification center is upright, while excelling in thermal resistance, on the occasion of resinification with formaldehyde, as for the hardened material, crosslinking density became high extremely, and it was very hard, and when the molding material was used as industrial use parts, there were [it was weak, and] restrictions.

[0004] Therefore, the present condition is having not resulted, by the time it acquires sufficient effect, although the attempt which denaturalizes melamine resin with rubber or denaturalizes it by phenol is made when using as a molding material. Moreover, although adding various alcohol, a saccharide, etc. into a melamine resin constituent, and improving crack-proof nature and a mechanical strength was performed, when copolycondensation was not enough at the time of manufacture, with time, the additive carried out the bleed out and there was a fault which causes the fall of a mechanical strength, the fall of crack-proof nature, a surface glossiness fall, etc.

[0005] Moreover, although it is inferior to the bendability of a paint film, and post-processing nature, therefore guanamines, such as benzoguanamine and acetoguanamine, may be used while it excels in thermal resistance, weatherability, and scratch-proof nature, although melamine resin is printed mainly as a curing agent of an alkyd resin and is used for a paint, since a benzoguanamine resin has a benzene machine in the structure, it is known that lightfastness is bad and a use has restrictions. Moreover, although alkoxy ** of the melamine-formaldehyde reactant was further carried out in alcohol, such as a methanol, ethanol, propanol, an isopropanol, and a butanol, and it was made to dissolve in a solvent when a melamine formaldehyde resin did not have the solubility over an organic solvent, therefore it used as an organic-solvent system paint, since a reaction process's becoming long and alkoxy ***** were acid and performed, condensation progressed and there was [a fault like control of molecular weight comes to spread in difficulty].

[0006] Moreover, melamine resin is conventionally used mainly from the level makeup side of furniture by a high degree of hardness, high thermal resistance, and high gloss as makeup material. Since design nature is increased in recent years, the so-called post form processing which carries out post processing of the panel after hardening is carried out briskly. Moreover, while a melamine facing plate has a high

degree of hardness, generating of a crack with time may be seen and the improvement is performed. [0007] Therefore, although improvement, improvement of a crack-proof, etc. were performed in post form processability by denaturalizing to melamine resin with the compound of various copolycondensation nature, or adding various additives to it, inclusion to a resin became poor with the waterproof fall and the fall of gloss, and was not necessarily satisfactory.

[0008] Moreover, the melamine is well used as woody system adhesives combining the urea. Although excelled in the productivity by a cure rate being early as compared with the phenol system adhesives used for the of-the-same-kind use, since the melamine system adhesives itself were lacking in lipophilic property, there was disadvantage to many [for the tar used / tar / suddenly in recent years] needle-leaf tree.

[0009]

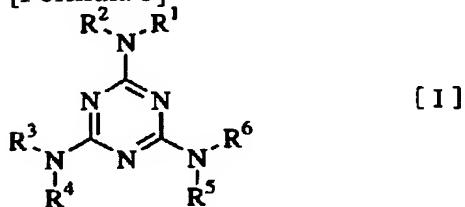
[Problem(s) to be Solved by the Invention] this invention offers the amino resin constituent which gives tough nature and bending nature and lipophilic property, without spoiling properties, such as the thermal resistance of melamine resin, a high degree of hardness, and outstanding gloss.

[0010]

[Means for Solving the Problem] this invention is a formula [I].

[0011]

[Formula 3]



[0012] A substituent, R1, R2, R3, R4, R5, and R6 express among [formula the substituent which became independent mutually respectively. 1 of these substituents or five pieces The alkyl group of C1-20, the alkenyl machine of C2-20 (this alkyl group or an alkenyl machine) You may have alicyclic structure or the phenyl group arbitrarily in the structure. moreover, when these two alkyl groups or an alkenyl machine is on the same nitrogen atom, or it may become together with the nitrogen atom and may form 3 - 8 member ring, a phenyl group is expressed and the remaining substituent expresses a hydrogen atom] It is related with the amino resin constituent manufactured by making formaldehyde react to one sort or two sorts or more of mixture of a triazine derivative which is alike and is expressed more.

[0013] Furthermore, this invention relates to the amino resin constituent manufactured by making formaldehyde react to the mixture which consists of one sort or two sorts or more of the mixture and melamines, or ureas of a triazine derivative which are expressed by the formula [I].

[0014] And this invention relates to the amino resin constituent containing the amino resin constituent manufactured by making formaldehyde react to one sort or two sorts or more of mixture of a triazine derivative expressed by the formula [I] and a melamine formaldehyde resin, a melamine and a formaldehyde resin, or a formaldehyde resin further. Hereafter, this invention is explained in detail.

[0015]

[The operation gestalt of invention] The substitution triazine derivative used for manufacture of the amino resin constituent of this invention 73 : (J. Am.Chem.Soc.), for example, the journal OBU American chemical society, easily obtained by the following well-known synthetic method and the same method, 2984 pages, the method of making the 2-chloro-1,3,5-triazine derivative and alkylamine of a publication react in 1951, and acquiring; 18 HEMISHIE BERIHITE (Chem Ber.), 2755 pages, How to make the 2, 4, and 6-trimethyl thio-1,3,5-triazine derivative and alkylamine of a publication react in 1885, and acquire; the 2, 4, and 6-triamino-1,3,5-triazine and the alkylamine hydrochloride of a publication to U.S. Pat. No. 2,228,161 (1941) method; German patent octavus No. 89,593 which is made to react and is obtained How to make the cyano piperidine and cyanoguanidine of a publication react to (1953), and to obtain 2-piperidino -4 and 6-diamino-1,3,5-triazine; the cyanuric chloride of a

publication, and corresponding alkylamines to JP,3-215564,A The method of making react and acquiring etc. is mentioned.

[0016] outside -- a method -- ***** -- this invention -- an applicant -- patent application -- carrying out -- having had -- the following -- a method -- it is -- : -- (-- a --) -- JP,8-27128,A -- a publication -- 1,3,5-triazine -- a derivative -- a periodic table -- the -- VII the [a group or] -- the bottom of existence of a VIII catalyst, alcohol, and method; made to react

(b) the [given in JP,8-193071,A 1,3,5-triazine derivative, and periodic-table] -- VII the [a group or] -- the [given in method; and (c) JP,8-27125,A which are made to react with aldehyde under existence of VIII catalyst and hydrogen content gas 1,3,5-triazine derivative, and periodic-table] -- VII the [a group or] -- an olefin, the method of making it react, etc. are mentioned under existence of a VIII catalyst, and a carbon monoxide / hydrogen mixed gas

[0017] The triazine derivative used for manufacture of the amino resin constituent of this invention may be a triazine derivative which is manufactured by which the above-mentioned method and to obtain. However, when using it for an electronic material field, the minute thing or the thing which is not ** of the adding-water resolvability chlorine which remains in substitution triazine is desirable. Therefore, the triazine derivative obtained by the method of making alcohols reacting to the melamine of the above-mentioned reference (a) under specific catalyst existence, the method of making aldehydes react to the melamine of (b) under specific catalyst existence in hydrogen atmosphere, or the method of making olefins react to the melamine of (c) under specific catalyst existence in hydrogen / carbon monoxide atmosphere is desirable.

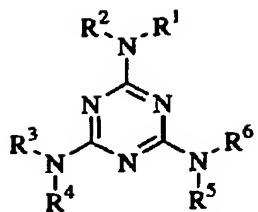
[0018] The alkyl group of C1-20, the alkenyl machine of C2-20 (this alkyl group or an alkenyl machine) You may have alicyclic structure or the phenyl group arbitrarily in the structure. Moreover, the substituent in the case of being expressed with a phenyl group, or it may become together with the nitrogen atom and may form the nitrogen-containing heterocycle of 3 - 8 member, when these two alkyl groups or an alkenyl machine is on the same nitrogen atom, R1, R2, R3, R4, and R5 And R6 : which carries out the following of the example -- a methyl group -- An ethyl group, n-propyl group, an isopropyl group, n-butyl, an iso-butyl, A sec-butyl, tert - A butyl, a cyclopropyl methyl machine, A cyclobutyl, n-pentyl machine, an iso-pentyl machine, a sec-pentyl machine, tert - A pentyl machine, a cyclopentylic group, n-hexyl machine, a cyclohexyl machine, A cyclohexyl methyl group, 4-methyl cyclohexyl methyl group, n-octyl machine, A 2-ethylhexyl machine, n-nonyl machine, n-decyl group, n-dodecyl, An n-hexadecyl machine, n-octadecyl machine, a benzyl, 1-phenethyl machine, 2-phenethyl machine, 1-phenylpropyl machine, 3-phenylpropyl machine, vinyl-group, allyl group, metallyl machine, clo chill machine, 2-pentenyl machine, 3-hexenyl machine, styryl machine, and phenyl group; -- and As a ring structure at the time of two substituents on the same nitrogen atom becoming together with the nitrogen atom, and forming a heterocycle, an aziridine ring, an azetidine ring, a pyrrolidine ring, a piperidine ring, etc. are mentioned. In addition, the range of the triazine derivative used by instantiation of these substituents for manufacture of the amino resin constituent of this invention is not limited.

[0019] In this invention, it is possible to use for the reaction of a reaction with formaldehyde, a melamine or a urea, and formaldehyde combining one sort of the triazine derivative expressed by the above-mentioned formula [I] or two sorts or more. The combination of the substituent shows still more concretely the main things of the triazine derivative used for the following table 1 by this invention.

[0020] In addition, :Me which shows the meaning of the following [cable address / of the substituent of front Naka] : A methyl group Et: Ethyl-group Pr: Normal propyl group ME:isopropyl machine Bu : [Normal butyl] IB: Isobutyl machine MP: Secondary butyl TB:tertiarybutyl machine PE: Normal pentyl machine HE: Normal hexyl machine EH:2-ethylhexyl machine Oc: normal octyl machine De : [Normal decyl group] DD -- :normal dodecyl HD: normal hexadecyl machine OD: normal octadecyl -- machine AL:allyl group ST:styryl machine Cy:cyclohexyl machine CM:cyclohexyl methyl group Ph: -- phenyl group Bz: -- benzyl

[0021]

[Formula 4]



[1]

[0022]

[Table 1]

Table 1 ----- R1 R2 R3 R4 R5 R6 ----- Me H H H HMe Me H H H HMe
 H Me H H HMe Me Me H H HMe H Me H MeHMe Me Me H HMe MeMe H Me HMe MeMe
 MeMe HEt H H H H HEt Et HHH HEt H Et HH HEt Et EtH H HEt HEt H Et HEt Et Et H HEt Et Et
 HEt HET Et Et Et Et HPr H H H H HPr Pr H H H HPr H Pr H H HPr Pr Pr H H HPr H PrH Pr HPr Pr
 PrH Pr H -----, [0023]

[Table 2]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- Pr Pr Pr Pr Pr HME H
 H H H HME ME H H H HME H ME H H HME ME ME H H HME H ME H ME HME MEME MEH
 HME MEME H ME HME ME ME ME ME HBu H H H H HBu Bu HHH HBu H Bu HH HBu Bu BuH
 H HBu HBu H Bu HBu Bu Bu BuH HBu Bu Bu H Bu HBu Bu Bu Bu HIB H H H H HIB IB HH H
 HIB H IBH H HIB IB IBH H HIB H IB HIB H ----- [0024]

[Table 3]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- IB IB IB IB H HIB IB
 IB H IB HIB IB IB IB HMP H H H H HMP MP H H H HMP H MP H H HMP MPMP H H HMP H
 MP H MP HMP MP MP MP H HMP MP MP H MP HMP MP MP MP MP HTB H H H H HTB TB
 HHH HTB H TB HH HTB TB TB H H HTB H TB H TB HTB TB TB TB H HTB TB TB H TB HTB
 TB TB TB TB HPE H H H HHPE PE HH H HPE H PE H HH----- [0225]

[Table 4]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- PE PE PE H H HPE H
PE H PE HPE PE PE PE H HPE PE PE H PE HPE PE PE PE PE HHE H H H H HHE HEH H H HHE
H HE H H HHE HE HE H H HHE H HE HHE HHE HE HEHE H HHE HE HE H HE HHE HE HE HE
HE HEH H H H H HEH EH HH HHEH H EH H H HEH EH EH H H HEHH EH HEH HEH EH EH
EH H HEH EH EH H EH HEH EH EH EH EH HOc H H H H H ----- [0026]

[Table 5]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- Oc Oc H H H HOc H
 Oc H H HOc Oc Oc H H HOc H Oc H Oc HOc Oc Oc H HOc Oc H Oc HOc OcOc OcOc HDe
 H H H H HDe De H H H HDe HH HDe De De H H HDe H De H De HDe De DeDe H HDe De De
 H De HDe De De DeHDD H H H H HDD DD HH H HDD H DD HH HDD DD DDH H HDD H
 DD H DD HDD DD DD DD H HDD DD DD H DD H ----- [0027]

[Table 6]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD DD DD DD DD
 HHD H H H H HHD HD H H H HHD H HD H H HHD HD HD H H HHD H HD H HD HHD HDHD
 HDH HHD HDHD H HD HHD HD HD HD HOD H H H HOD OD HHH HOD H OD HH HOD
 OD ODH H HOD HOD H OD HOD OD OD ODH HOD OD OD H OD HOD OD OD OD OD HAL H
 H H H HAL AL HH H HAL H ALH H HAL AL ALH H HAL H AL HAL H----- [0028]

[Table 7]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- Bz H Bz H Bz HDD
 Me H H H HDD H Me H H HDD Me Me H H HDD H Me H MeHDD Me Me H H HDD MeMe H Me
 HDD MeMe MeMe HDD Et H H H HDD H Et H H HDD Et EtH H HHD H Et HEt HDD Et Et
 HHDD Et EtH Et HDD Et Et Et HDD Pr H H H HDD H Pr H H HDD Pr Pr H H HDD H Pr HPr
 HDD PrPr HPr HDD Pr Pr Pr Pr HDD ME H H H H ----- [0030]

[Table 9]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD H ME H H HDD
 ME ME H H HDD H ME H ME HDD ME ME ME H HDD ME ME H ME HDD ME ME ME
 HDD BuH H H HDD H Bu H H HDD Bu Bu H H HDD H Bu HBu HDD Bu BuH HDD Bu Bu H
 Bu HDD Bu Bu Bu HDD IB H H H HDD H IB H H HDD IB IB H H HDDH IB H IB HDD IB IB
 IBH HDD IB IB H IB HDDIB IB IB HDD MP H H H HDD H MP H HH ----- [0031]

[Table 10]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD MP MP H H HDD
 H MP H MP HDD MP MP MP H HDD MP MP H MP HDD MP MP H MP HDD TB H H H HDD H
 TB H H HDD TBTB H H HDD H TBH TB HDD TB TB H HDD TB TB HTB HDD TB TB TB
 HDD PE H HH HDD H PE H H HDD PE PE H HHDD H PE H PE HDD PE PEPE H HDD PE PE H
 PE HDD PE PE PE HDD HE H H H HDD H HE H H HDD HE HEH H H ----- [0032]

[Table 11]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD H HE H HE HDD
 HE HE HE H HDD HE HE H HE HDD HE HE HE HDD EH H H H HDD H EH H H HDD EHEH
 H H HDD H EH H EH HDD EH EH EH HHDD EH EH HEH HDD EH EH EH HDD Oc H HH
 HDD H Oc H H HDD Oc Oc H HHDD HOc H Oc HDD Oc Oc H HDD Oc Oc H Oc HDD Oc Oc
 Oc Oc HDD De H H H HDD H De H H HDD De DeH H HDD HDe H De H ----- [0033]

[Table 12]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD De De De H HDD
 De De H De HDD De De De HDD HD H H H HDD H HD H H HDD HD H H HDD H HD H
 HD HDD HDHD HDH HDD HD HD H HD HDD HD HD HD HDD OD HH H HDD H OD H H
 HDDOD OD HHDD H OD HOD HDD OD OD OD H HDD OD OD H OD HDD OD OD OD OD
 HDD AL H H H HDD H AL H H HDDAL AL HH HDD H AL HAL HDD AL AL AL H H -----
 [0034]

[Table 13]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- DD AL AL H AL
 HDD AL AL AL AL HDD ST H H H HDD H ST H H HDD ST ST H H HDD H ST H ST HDD STST
 STH HDD STST H ST HDD ST ST ST HDD H Cy H H HDD H Cy H Cy HDD HCM HH HDD
 HCM H CM HDD H Ph H H HDD H Ph H Ph HDD H Bz H H HDD H Bz H Bz HHD Me H H H HHD
 HMe H HHHD Me Me HH HHD H Me H Me HHD Me Me Me H H ----- [0035]

[Table 14]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD Me Me H Me
 HHD Me Me Me HHD Et H H H HHD H Et H H HHD Et Et H H HHD H Et H Et HHD EtEt EtH
 HHD EtEt H Et HHD Et Et Et HHD Pr H H H HHD H Pr H H HHD Pr PrH H HHD H Pr HPr HHD
 Pr Pr HPr HHD Pr Pr Pr Pr HHD ME H H H HHD H ME H H HHD ME ME H H HHD HME H ME
 HHD ME ME ME H HHD ME ME H ME HHD ME ME ME H ----- [0036]

[Table 15]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD Bu H H H HHD H
 Bu H H HHD Bu Bu H H HHD H Bu H Bu HHD Bu Bu Bu H HHD Bu Bu H Bu HHD BuBu BuBu
 HHD IBH H H HHD H IBH H HHD IB IBH H HHD H IB HIB HHD IB IB HHHD IB IBH IB HHD
 IB IB IB HHD MP H H HHHD H MP H H HHD MP MPH H HHD H MP HMP HHD MP MPMP
 HHHD MP MPH MP HHD MP MP MP HHD TB H H H H ----- [0037]

[Table 16]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD H TB H H HHD
 TB TB H H HHD H TB H TB HHD TB TB H HHD TB TB H TB HHD TB TB H HHD PEH
 H H HHD H PE H H HHD PE PE H H HHD H PE HPE HHD PE PE PEH HHD PE PE H PE HHD PE
 PE PE PE HHD HE H H HHD H HE H H HHD HE HE H H HHD HE H HE HHD HE HE HE HEH
 HHD HE HE H HE HHD HE HE HE HHD EH H H H HHD H EH H HH----- [0038]

[Table 17]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD EH EH H H HHD
 H EH H EH HHD EH EH EH H HHD EH EH EH EH EH EH HHD Oc H H H HHD H Oc
 H H HHD Oc Oc H H HHD H Oc H Oc HHD Oc Oc Oc H HHD Oc Oc Hoc HHD Oc Oc Oc Oc HHD
 De H HH HHD H De H H HHD De De H HHHD H De H De HHD De De De H HHD De De H De
 HHD De De De HHD DD H H H HHD H DD H H HHD DD DDH H H----- [0039]

[Table 18]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD H DD H DD HHD
 DD DD DD H HHD DD DD H DD HHD DD DD DD HHD OD H H H HHD H OD H H HHD
 ODOD H H HHD H OD H OD HHD OD OD OD HHHD OD OD HOD HHD OD OD OD OD HHD
 AL H HH HHD H AL H H HHD AL AL H HHHD HAL H AL HHD AL AL AL H HHD AL AL H AL
 HHD AL AL AL AL HHD ST H H HHD H ST H H HHD ST STH H HHD HST H ST H-----
 ----- [0040]

[Table 19]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- HD ST ST ST H HHD
 ST ST H ST HHD ST ST ST HHD H Cy H H HHD H Cy H Cy HHD H CM H H HHD H CM H
 CM HHD H Ph H H HHD H Ph H Ph HHD H Bz H H HHD H Bz H Bz HOD Me H H H HOD H Me
 HH HOD Me Me HH HOD H Me HMe HOD Me Me Me H HOD MeMe H Me HOD Me Me Me Me
 HOD Et HH H HOD H Et H H HOD Et Et HH HOD HEt H Et H----- [0041]

[Table 20]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- OD Et Et Et H HOD
 Et Et H Et HOD Et Et Et HOD Pr H H H HOD H Pr H H HOD Pr Pr H H HOD H Pr H Pr HOD PrPr
 H Pr HOD Pr Pr Pr HOD ME H H H HOD H ME H H HOD ME ME H H HOD HME HME HOD
 ME ME MEH HOD ME ME H ME HOD ME ME ME HOD Bu H H H HOD H Bu H H HOD Bu
 Bu H H HOD H Bu HBu HOD Bu Bu Bu H HOD Bu Bu H Bu H Bu H----- [0042]

[Table 21]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- OD Bu Bu Bu Bu
 HOD IB H H H HOD H IB H H HOD IB IB H H HOD IB IB H H HOD IBIB H IB
 HOD IBIB IBIB HOD MP H H H HOD H MP H H HOD MP MPH H HOD H MP HMP HOD MP MP
 MP HHOD MP MPH MP HOD MP MP MP HOD TB H H H HOD H TB H H HOD TB TB H H
 HOD H TB H TB HOD TB TB H HOD TB TB H TB HOD TB TB TB H-----
 [0043]

[Table 22]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- PE PE H H H HOD H
 PE H H HOD PE PE H H HOD H PE H PE HOD PE PE PE H HOD PE PEPE PEPE
 HOD HEH H H HOD H HEH H HOD HE HEH H HOD H HE HHE HOD HE HE HE HHOD HE HEH
 HE HOD HE HE HE HOD EH H H HHOD H EH H H HOD EH EHH H HOD H EH HEH HOD
 EH EHEH HHOD EH EHH EH HOD EH EH EH HOD Oc H H H H----- [0044]

[Table 23]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- OD H Oc H H HOD
 Oc Oc H H HOD H Oc H Oc HOD Oc Oc Oc H HOD Oc Oc Oc Oc HOD DeH H H
 HOD H De H H HOD De De H H HOD H De HDe HOD De De DeH HOD De De H De HOD De De
 De De HOD DD H H H HOD H DD H H HOD DD DD H H HODH DD H DD HOD DD DD DDH
 HOD DD DD H DD HODDD DD DD DD HOD HD H H H HOD H HD H HH-----
 [0045]

[Table 24]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- OD HD HD H H HOD H HD H HD HOD HD HD H HOD HD HD H HD HOD HD HD H HD HOD AL H H H HOD H AL H H HOD ALAL H H HOD H ALH AL HOD AL AL AL H HOD AL AL HAL HOD AL AL AL AL HOD ST H HH HOD H ST H H HOD ST ST H HHOD H ST H ST HOD ST STST H HOD ST ST H ST HOD ST ST ST HOD H Cy H H HOD H Cy H H CM HH H-----.

[0046]

[Table 25]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- OD H CM H CM HOD H Ph H H HOD H Ph H Ph HOD H Bz H H HOD H Bz H Bz HMe H EH H H HMe MeEH H H HMe H EH H Me HMe Me EH H Me HMe Me Me EHHMe H Cy HH HMe Me Cy H H HMe H Cy H Me HMe Me CyH Me HMe Me MeMe Cy HMe H Bz H H HMe Me Bz H H HMe H Bz H Me HMe Me Bz H Me HMe Me Me Bz HEt H EH H H HEt Et EH H H H-----. [0047]

[Table 26]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- Et H EH H Et HEt Et EH H Et HEt Et Et EH HEt H Cy H H HEt Et Cy H H HEt H Cy H Et HEt EtCy H Et HEt EtEt EtCy HEt H Bz H H HEt Et Bz H H HEt H Bz H Et HEt Et Bz H Et HEt Et Et Bz HME H EH HH HME H Cy HH HME H Cy H ME HME H Bz HH HME H Bz H ME HBu H EH HH HBu Bu EH HH HBu H EH H Bu HBu Bu EH H Bu H-----. [0048]

[Table 27]

Table 1 It continues. ----- R1 R2 R3 R4 R5 R6 ----- Bu Bu Bu Bu EH HBu H Cy H H HBu Bu Cy H H HBu H Cy H Bu HBu Bu Cy H Bu HBu Bu Bu Cy HBu H Bz H H HBu BuBz H H HBu H BzH Bu HBu Bu Bz H Bu HBu Bu Bu Bz HAL H EH HH HAL AL EH H H HAL H EH H AL HAL AL EH H AL HAL AL AL EH HAL HCy H HHAL AL Cy H H HAL H Cy H AL HAL AL Cy H AL HAL H Bz H H HAL AL Bz H H H-----. [0049]

[Table 28]

表1 続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
AL	H	Bz	H	AL	H
AL	AL	Bz	H	AL	H
AL	AL	AL	AL	Bz	H
-(CH ₂) ₄ -		H	H	H	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -		H	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -	Bu	H	
-(CH ₂) ₆ -		H	H	H	H
-(CH ₂) ₆ -		H	H	Me	H
-(CH ₂) ₆ -		H	H	Et	H
-(CH ₂) ₆ -		H	H	Pr	H
-(CH ₂) ₆ -		H	H	Bu	H
-(CH ₂) ₆ -		-(CH ₂) ₆ -		H	H
-(CH ₂) ₆ -		-(CH ₂) ₆ -	Bu	H	
-(CH ₃) ₄ -		EH	H	H	H
-(CH ₂) ₄ -		EH	H	Bu	H
-(CH ₂) ₄ -		EH	H	MP	H
-(CH ₃) ₄ -		EH	H	PE	H
-(CH ₃) ₄ -		EH	H	HE	H
-(CH ₂) ₄ -		-(CH ₂) ₄ -		EH	H
-(CH ₂) ₄ -		Cy	H	H	H
-(CH ₂) ₄ -		Cy	H	IB	H
-(CH ₂) ₄ -		Cy	H	EH	H

[0050]
 [Table 29]

表1 続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
-(CH ₂) ₄ -	C y	H	O c	H	
-(CH ₂) ₄ -	C y	H	O D	H	
-(CH ₂) ₄ -		-(CH ₂) ₄ -	C y	H	
-(CH ₂) ₄ -	B z	H	H	H	
-(CH ₂) ₄ -	B z	H	M e	H	
-(CH ₂) ₄ -	B z	H	E t	H	
-(CH ₂) ₄ -	B z	H	P r	H	
-(CH ₂) ₄ -	B z	H	B u	H	
-(CH ₂) ₄ -		-(CH ₂) ₄ -	B z	H	
-(CH ₂) ₅ -	E H	H	H	H	
-(CH ₂) ₅ -	E H	H	M e	H	
-(CH ₂) ₅ -	E H	H	E t	H	
-(CH ₂) ₅ -	E H	H	P r	H	
-(CH ₂) ₅ -	E H	H	E H	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	E H	H	
-(CH ₂) ₅ -	C y	H	H	H	
-(CH ₂) ₅ -	C y	B u	H	H	
-(CH ₂) ₅ -	C y	H E	H	H	
-(CH ₂) ₅ -	C y	H	D e	H	
-(CH ₂) ₅ -	C y	H	H D	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	H	H	
-(CH ₂) ₅ -	B z	H	H	H	

[0051]

[Table 30]

表1 続き

R ¹	R ²	R ³	R ⁴	R ⁵	R ⁶
-(CH ₂) ₅ -	B z	D e	H	H	
-(CH ₂) ₅ -	B z	H D	H	H	
-(CH ₂) ₅ -	B z	H	D e	H	
-(CH ₂) ₅ -	B z	H	H D	H	
-(CH ₂) ₅ -		-(CH ₂) ₅ -	B z	H	

[0052] The inside of the combination of the substituent of the above-mentioned formula [I] and a desirable thing are a substituent, R1, R2, R3, R4, and R5. And R6 Inside, R1, and R3 And R5 It is the group to which 1 or three pieces express the alkyl group of C1-10, and the remaining substituent expresses a hydrogen atom.

[0053] A still more desirable thing in the combination of the substituent of the above-mentioned formula [I] A substituent, R1, R2, R3, R4, and R5 And R6 Inside, R1 and R3 And R5 It is the group to which 1

or three pieces express a methyl group, an ethyl group, n-propyl group, an isopropyl machine, n-butyl, a cyclohexyl machine, or a 2-ethylhexyl machine, and the remaining substituent expresses a hydrogen atom.

[0054] In the reaction of the one sort or two sorts or more of mixture and formaldehyde of a triazine derivative which are expressed by the formula [I] for manufacturing an amino resin constituent, the number of mols of the formaldehyde to one mol of one sort or two sorts or more of mixture of a triazine derivative has the desirable number of mols of the non-replaced hydrogen atom of the amino group of a triazine derivative, and the same number from one mol to one mol of triazine derivatives. If there are few mols of formaldehyde than 1, the structure generated according to heat curing of a resin constituent cannot form the structure of cross linkage which becomes enough, and cannot satisfy physical properties, such as the thermal resistance of the heat-curing object of an amino resin constituent, a high degree of hardness, and toughness. If [than the number of mols of the non-replaced hydrogen atom of the amino group of an another side triazine derivative, and the same number] more, the formaldehyde which was not able to be kept to a triazine derivative and an addition reaction remains. and removal of the formalin which remains spreads in difficulty -- therefore, the formalin smell of an amino resin constituent -- ***** -- it is not desirable The formalin smell in the case of heat curing is also remarkable. Moreover, use of superfluous formaldehyde is economically disadvantageous.

[0055] Although the reaction of the triazine derivative of this invention and formaldehyde is usually performed in an aquosity medium, since the triazine derivative of this invention is meltable to usually and an organic solvent, if a meltable organic solvent is added also to water, a reaction with formaldehyde will progress smoothly. As an organic solvent, polar solvents, such as ether, such as alcohols [, such as a methanol ethanol, an isopropanol, and propanol,], dioxane, tetrahydrofuran, 1, and 2-dimethoxyethane, a dimethylformamide, and dimethyl sulfoxide, are desirable.

[0056] In the resinification reaction which is a reaction of a triazine derivative and formaldehyde, reaction temperature is usually within the limits of 50-95 degrees C. However, when a reaction may advance also at low temperature from this range, for example, an organic solvent is added, a resinification reaction advances at low temperature from 50 degrees C.

[0057] pH of the reaction mixture of resinification reaction time -- 6-10 -- it is 7-9 suitably In order to increase the molecular weight of the product by the resinification reaction, pH is 7-7.5 a neutrality side, and when stopping molecular weight low, it advances a resinification reaction by the alkali side, pH 8-9 [for example,].

[0058] Reaction time, i.e., time until it results [from a resinification reaction start] in a reaction end, is decided by considering the time of the resin constituent of desired molecular weight being obtained as a reaction end time. Molecular weight is suitably sampled among a reaction and is determined by the determination of molecular weight by the nebula temperature method, the degree method of water mixing, and/or the gel par MYUESHON chromatography method. As mentioned above, in this invention, making one sort or two sorts or more of mixture of a triazine derivative mix a melamine or a urea, and resinifying is also performed in the reaction of the one sort or two sorts or more of mixture and formaldehyde of a triazine derivative which are expressed by the formula [1].

[0059] Although crosslinking density will become high and a degree of hardness will increase if the amount of the melamine generally made to contain or a urea becomes large, although there is no limit of an amount to the triazine derivative of the melamine or urea which one sort or two sorts or more of mixture of a triazine derivative is made to mix, it becomes weak and tough nature falls. 20 mols of molar quantity of the melamine made to contain or a urea are ten mols from 0.1 mols from 0.05 preferably to one mol of triazine derivatives.

[0060] The reaction condition in the reaction of one sort or two sorts or more of mixture of a triazine derivative expressed to the formula [I] mixed with the melamine or the urea and formaldehyde, i.e., a solvent, reaction temperature, pH of reaction mixture, reaction temperature, etc. are the same as them of the reaction of the triazine derivative and formaldehyde which were mentioned above. The number of mols of the formaldehyde in this reaction has the desirable number of mols of the non-replaced hydrogen atom of the amino group of a melamine, a urea, and a triazine derivative, and the same number

from the equal number of mols to the number of sum total mols of a melamine, a urea, and a triazine derivative. When fewer than the equal number of mols, bridge formation cannot fully form and physical properties are inferior. the non-replaced hydrogen atom of the amino group of an another side melamine, a urea, and a triazine derivative, and the formaldehyde which was not able to be kept to an addition reaction or a condensation reaction with the amino group when [than the number of mols of the same number] more -- remaining -- moreover, this removal -- difficulty -- carrying out -- hurting -- a constituent -- a formalin smell -- ***** -- it is not desirable Moreover, use of superfluous formaldehyde is economically disadvantageous.

[0061] Moreover, using mixture with the amino resin constituent which one sort or two sorts or more of the mixture and formaldehyde of a triazine derivative which are expressed by the formula [1] of this invention are made to react, and is obtained, a melamine formaldehyde resin, a melamine and a formaldehyde resin, or a formaldehyde resin is also performed. Mixture means the thing which comes to mix the liquefied article of the liquefied article of an amino resin constituent or solid-state-like article which one sort or two sorts or more of the mixture and formaldehyde of a triazine derivative of this invention are made to react, and is obtained, at least one sort of melamine formaldehyde resins chosen from from while being obtained by the well-known method, a melamine and a formaldehyde resin, or a formaldehyde resin, or the solid-state-like article of that at an arbitrary rate.

[0062] It is the with 55% of solid-content concentration, and a molecular weight of about 600 resin liquid which one mol of melamines and 1.6 mols of formaldehyde were made to react temperature about 60 minutes of 95 degrees C, and obtained it by pH eight as instantiation of the melamine formaldehyde resin obtained by the well-known method. Moreover, it is the with 55% of solid-content concentration, and a molecular weight of about 700 resin liquid which 0.2 mols of melamines, one mol of ureas, and 1.8 mols of formaldehyde were made to react pH 7.9 temperature of 90 degrees C, and about 120 minutes, and was obtained as instantiation of a melamine and a formaldehyde resin. Furthermore, it is the with 50% of solid-content concentration, and a molecular weight of about 500 resin liquid which one mol of ureas and 1.8 mols of formaldehyde were made to react pH 8.0 temperature of 90 degrees C, and about 150 minutes, and was obtained as instantiation of a urea-resin again.

[0063] Although the mixed rate of a triazine derivative-formaldehyde resin constituent, the aforementioned melamine formaldehyde resin, a melamine and a formaldehyde resin, and a formaldehyde resin is the thing of the large range, when the rate of a triazine derivative-formaldehyde resin constituent is high, the improvement effect given to tough nature and bending nature, lipophilic property, etc. is large, and if a rate is low, on the other hand, the improvement effect will become scarce. although loadings change with a use and purposes -- the triazine derivative-formaldehyde resin constituent 100 weight section -- receiving -- a melamine formaldehyde resin, a melamine and a formaldehyde resin, or a formaldehyde resin -- the 20 - 1000 weight section -- it is the 50 - 500 weight section suitably

[0064] The amino resin constituent manufactured as mentioned above is used for fields, such as woody material adhesives, forming material, makeup material, and a paint. When used as adhesives of a woody material, a curing agent is mixed, on wood, it mixes with an application or wood flour, and an amino resin constituent heat-hardens by the hot press. A thickener, an osmosis control agent, etc. are added if needed in that case.

[0065] While adding a curing agent, mixing with wood flour, pulp powder, inorganic powder, etc. and carrying out type with a twist to interweaving by heating under pressure after making it dryness powder while it has been liquefied or when used as forming material, an amino resin constituent heat-hardens. When used as makeup material, paper etc. sinks in and dries, the amino resin constituent resin liquid which added the curing agent is combined with a phenol resin impregnated paper or a woody board, and an amino resin constituent heat-hardens by the hot press.

[0066] When used for a paint, an amino resin constituent is combined with an alkyd resin, polyester resin, acrylic resin, etc., adds a curing agent and other additives, considers them as paint composition, and is used as OH basis of the resin combined by heating, and a cross linking agent which reacts. The demand gestalt of a paint considers as resins, such as an aqueous solution type, an organic-solvent

dissolution type, and a solid type, in that case.

[0067]

[Example] Although an example and the example of comparison explain this invention still in detail below, the publication of these examples does not mean limitation of the range of this invention. The triazine derivative used for an example was compounded by the method of the following examples of reference.

Composition of the example 1:2 of reference, and 4-diamino-6-normal butylamino-1,3,5-triazine. [0068]
 (a) Stirring violently, 303.7g (5.0 mols) of 28% aqueous ammonia solution was dropped at the solution which cooled 184.5g (1.0 mols) of synthetic cyanuric chloride of 2 and 4-diamino-6-crawl-1,3,5-triazine at 0 degree C after the dissolution at the room temperature at acetonitrile 800mL in 2 hours so that 10 degrees C or less might be maintained for reaction temperature. After stopping cooling and stirring at a room temperature for 1 hour, it warmed gradually and was made to react as 45 degrees C after a dropping end for further 4 hours. The product was carried out the RO exception after cooling, and still a lot of water washed. 115g (79% of yield) of notation compounds was obtained by drying a filtration object at 50 degrees C under a vacuum for 6 hours.

[0069] (b) 2, 2 compounded by composition (a) of 4-diamino-6-normal butylamino-1,3,5-triazine, and 4-diamino-6-crawl-1,3,5-triazine 14.5g (0.1 mols), water 100mL, and the butylamine 29.2g (0.4 mols) mixed solution were warmed stirring, and were made to react at reflux temperature finally for 6 hours. The product was carried out the RO exception after cooling reaction mixture, still a lot of water fully washed, and, next, it washed with toluene. 17.5g (96% of yield) of declared compounds was obtained by drying a filtration object at 70 degrees C under a vacuum for 6 hours. Melting point; 167 degrees C.

[0070] example of reference 2: -- the autoclave equipped with 2, 4, the synthetic agitator of 6-tris (normal butylamino)-1,3,5-triazine, and the thermometer -- melamine 12.6g (0.1 mols) 1 and 4-dioxane 200g, n-butyraldehyde 72.0g (1.0 mols), and 2.0g of 5%Pd support activated carbon -- teaching -- after nitrogen gas substitution and hydrogen gas -- initial pressure 40 kg/cm² ** -- it carries out, and it was made to react with the reaction temperature of 180 degrees C for 6 hours, agitating violently After having cooled slowly to the room temperature after the reaction end, carrying out the RO exception and removing a catalyst and a solid, the solvent was distilled off and the rough object of a ***** liquefied resultant was obtained. After having made the acetone / hexane partially aromatic solvent into the eluate for this with the silica gel column chromatography, developing the mixed ratio, changing concentration into 100 / 1 - 1/100 serially and isolating a product, the solvent was distilled off and 19.6g of liquefied notation compounds was obtained.

[0071] The 2 and 4-diamino-6-crawl-1,3,5-triazine 14.5g (0.1 mols), water 140mL, and cyclohexylamine 29.2g [of the intermediate field compounded in the example 3:4 of reference and the example 1 of synthetic reference of 6-diamino-2-cyclohexylamino-1,3,5-triazine] (0.3 mols) mixed solution was warmed stirring, and was made to react at reflux temperature for 1 hour. Furthermore, the water 40mL solution of 12g of sodium hydroxides was dropped over 1 hour, and it riped for 1 hour. Toluene 200mL was added to the obtained reaction mixture, and it cooled to the room temperature after that. the obtained crystal -- separating -- toluene -- 17.9g (86% of yield) of notation compounds was obtained by drying under reduced pressure after washing one by one by water 100mL continuously 100 mL Melting point; 151 degrees C.

[0072] The example 4:4 of reference, 6-diamino -2 -(2-ethyl hexylamino)- It compounded like the example 3 of reference except changing to synthetic cyclohexylamine 29.2g (0.3 mols) of 1,3,5-triazine, and using 2-ethylhexyl amine 12.9g (0.1 mols). 18.6g (78% of yield) of ****. Melting point; 81 degrees C.

[0073] example of reference 5: -- 40% of 308g (4.0 mols) dropping of monomethylamine solution is carried out, agitating violently 184.5g (1.0 mols) of synthetic cyanuric chloride of 2, 4, and 6-tris (methylamino)-1,3,5-triazine in the solution cooled after the dissolution at the room temperature to toluene 920g at 0 degree C After holding 0 degree C after a dropping end for 2 hours, agitating, it warmed gradually and churning was performed as 30 degrees C for further 1 hour. The temperature up of the solution was carried out to 40 more degrees C, it agitated for 2 hours and the temperature up was

carried out to 70 degrees C. Holding the solution at 70 degrees C, 46g and 40% of 464g (6.0 mols) dropping of the dioxane of monomethylamine solution was carried out, and churning was continued for 9 hours. 300g (3.0 mols) was added for sodium-hydroxide solution to 30 degrees C after [cooling] 40%, and it agitated for 1 hour, and it cooled at 5 more degrees C, and agitated for 2 hours. The product was carried out the ** exception and washed with water. By drying a filtration object at 50 degrees C for 6 hours, 93g (55% of yield) was obtained for the notation compound. 133 degrees C of melting points.

[0074] 2 compounded by method of example 1 of example 1 reference and 4-diamino-6-normal butylamino-1,3,5-triazine 455g, 40% formalin 300g, 184.3g [of water], and dioxane 455g was taught to 2L4 mouth flask equipped with the agitator, the thermometer, and the cooling pipe, and pH was adjusted to 8.0 in KOH 5%. Then, agitating, carried out the temperature up, kept temperature at 70 degrees C, and it was made to react for 2 hours, and cooled to the room temperature after that.

[0075] Vacuum concentration was carried out at 40 degrees C, and 540g was obtained for the yellow transparent oil-like resin constituent. The molecular weight measured by the gel par MYUESHON chromatography method (the GPC method) was 580.

[0076] 2 obtained by method of publication for example 2 of example 2 reference, 4, and 6-tris (normal butylamino)-1,3,5-triazine 264.6g, 40% formalin 150g, 50g [of water], and dioxane 150g was taught to 2L4 mouth flask. Based on the example 1, 300g of oily brown resin constituents was obtained. The molecular weight measured by the GPC method was 520.

[0077] 4 obtained by method of publication for example 3 of example 3 reference and 6-diamino-2-cyclohexylamino-1,3,5-triazine 520g, 40% formalin 300g, 184.3g [of water], and dioxane 520g was taught to 2L4 mouth flask, and 589g was obtained for the oily resin constituent based on the example 1. The molecular weight by the GPC method was 720.

[0078] 4, the 6-diamino -2 which were obtained by the method of the example 4 of example 4 reference -(2-ethyl hexylamino)- 1,3,5-triazine 476g, 40% formalin 240g, 147.5g [of water], and dioxane 455g was taught to 2L4 mouth flask, and the wax-like resin constituent was obtained based on the example 1. The molecular weight by the GPC method was 670.

[0079] 2 compounded by method of example 5 melamine 63g and example 1 of reference and 4-diamino-6-normal butylamino-1,3,5-triazine 91g, 40% formalin 120g, 50g [of water], and dioxane 181g was taught to 1L4 mouth flask equipped with the agitator, the thermometer, and the cooling pipe, and pH was adjusted to 9.2 in NaOH solution 10%. Then, agitating, the temperature up was carried out, temperature was kept at 80 degrees C, and it was made to react for 90 minutes. It cooled to the room temperature after that, and transparent liquid was obtained. The molecular weight by the GPC method was 850.

[0080] 2 compounded by method of example 1 of example 6 reference and 4-diamino-6-normal butylamino-1,3,5-triazine 455g, 40% formalin 300g, 184.3g [of water], and dioxane 455g was taught to 2L4 mouth flask equipped with the agitator, the thermometer, and the cooling pipe, and pH was adjusted to 8.4 in KOH solution 5%. Then, agitating, the temperature up was carried out, temperature was kept at 70 degrees C, and it was made to react for 2 hours.

[0081] It cooled to the room temperature, pH was adjusted to 9.0 in KOH 5% after cooling, and resin liquid was obtained. 500g of this resin liquid and 500g (Nissan Chemical Industries shrine tradename SANTOPPU and M700 resin solid 55% per part) of melamine resin liquid were mixed. Resin liquid saves [for one month] at a room temperature and was stable.

[0082] 2 compounded by method of example 5 of example 7 reference, 4, and 6-tris (methylamino)-1,3,5-triazine 420g, 40% formalin 300g, and 360g of water were taught to 2L4 mouth flask, and it was made to react based on an example 1. The molecular weight by the GPC method of a product was 420.

[0083] Example of comparison 1 melamine 1260g, 40% formalin 1200g, and 1400g of water were taught to 5L4 mouth flask equipped with the agitator, the thermometer, and the cooling pipe, and pH was adjusted to 8.4 in KOH 10%. Then, agitating, the temperature up was carried out, temperature was kept at 95 degrees C, and it was made to react for 60 minutes. It cooled to the room temperature after that, pH was adjusted to 9.0 in KOH solution 10%, and the transparent water resin was obtained. The molecular weight measured by the GPC method was 680.

[0084] In addition, 30% acetone solution of Para toluenesulfonic acid was used as the hypoviscosity liquid at the resin liquid obtained in the example of evaluation 1 examples 1-4 to resin liquid 3% of the weight as a small amount of acetone and a curing agent. It sank into the chemistry filter paper (tradename [by Toyo Roshi Kaisha, Ltd.] 5C), the liquid was dried at 105 degrees C after that, the acetone was removed, and the impregnated paper which made 60% coating weight of a resin to the weight of a filter paper was obtained. Each resin liquid was infiltrated into the filter paper about examples 5 and 6 and the example 1 of comparison, and the impregnated paper which makes coating weight of a resin 60% after dryness at 105 degrees C was obtained.

[0085] each obtained impregnated paper -- a five-sheet laminating -- carrying out -- 150 degrees C and pressure 10 kg/cm² -- the heat press was carried out for 10 minutes, and the laminating hardening sheet with a thickness of 0.8mm was obtained. The laminating sheet was cut down for the strip-of-paper type sample with a length of 40mm, a width of face [of 25mm], and a thickness of 0.8mm, and it bent until it uprighted the strength-test machine (tradename [by the cage en tech company] tensilon UCT- 10 T) and fractured it by part for 5mm/in crookedness speed.

[0086] It is the original length L0 It is the length when fracturing by carrying out L1 It carries out and is Flexibility F F=(L0-L1) x100 / L0 It asked by carrying out. A result is shown in Table 2.

[0087]

[Table 31]

表2

	実施例						比較例
	1	2	3	4	5	6	
屈曲性							
F (%)	4 8	6 5	5 2	6 0	4 0	4 1	9

[0088] the resin liquid which mixed 200g of resin liquid obtained in the example of evaluation 2 example 7, and 800g (55% of tradename [by Nissan Chemical Industries, Ltd.] SANTOPPU M700 solid contents) of melamine resin liquid, and melamine resin liquid (55% of tradename [by Nissan Chemical Industries, Ltd.] SANTOPPU M700 solid contents) -- independently, it added 3% to resin liquid, and 30% solution of Para toluenesulfonic acid was used as the resin liquid for sinking in, respectively It is each resin liquid 80g/m² The tissue was infiltrated and the impregnated paper of 60% of coating weight of a resin was obtained after dryness at 105 degrees C.

[0089] each obtained impregnated paper -- a three-sheet laminating -- carrying out -- 150 degrees C and pressure 10 kg/cm² -- the hot press was carried out for 10 minutes, and the 0.5mm laminating hardening sheet was obtained the result which carried out bending for each laminating sheet at 120 degrees C -- melamine resin -- although 12mmR(s) or the crack occurred if independent, as for the object which mixed the resin liquid obtained in the example 7 20%, a crack did not occur in 6mmR(s), either

[0090] The flexibility and post form nature of a laminating hardening sheet which were obtained using the resin liquid of an example are notably higher than that melamine resin independent so that clearly from the result of the examples 1 and 2 of evaluation. As mentioned above, since the amino resin constituent of this invention has toughness and post form nature, without spoiling properties, such as the thermal resistance of melamine resin, a high degree of hardness, and outstanding gloss, it not only can use for a laminating sheet, a panel, a molding material, etc., but grant of organic-solvent solubility, fresh oil nature, etc. can use it for fields, such as adhesion of eye a possible hatchet, a paint, and difficulty adhesion wood, by choosing a substituent.

[Translation done.]

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PRIOR ART

[Description of the Prior Art] Melamine resin is conventionally used for a molding material, makeup material, the curing agent for paints, woody system adhesives, etc. widely for the reasons nil why mold releasability with metal mold is good etc., at the time of that the outstanding thermal resistance, a high degree of hardness, high gloss, and a cure rate are early and fabrication.

[0003] On the other hand, since melamines were that it is six functionality and that a triazine ossification center is upright, while excelling in thermal resistance, on the occasion of resinification with formaldehyde, as for the hardened material, crosslinking density became high extremely, and it was very hard, and when the molding material was used as industrial use parts, there were [it was weak, and] restrictions.

[0004] Therefore, the present condition is having not resulted, by the time it acquires sufficient effect, although the attempt which denaturalizes melamine resin with rubber or denaturalizes it by phenol is made when using as a molding material. Moreover, although adding various alcohol, a saccharide, etc. into a melamine resin constituent, and improving crack-proof nature and a mechanical strength was performed, when copolycondensation was not enough at the time of manufacture, with time, the additive carried out the bleed out and there was a fault which causes the fall of a mechanical strength, the fall of crack-proof nature, a surface glossiness fall, etc.

[0005] Moreover, although it is inferior to the bendability of a paint film, and post-processing nature, therefore guanamines, such as benzoguanamine and acetoguanamine, may be used while it excels in thermal resistance, weatherability, and scratch-proof nature, although melamine resin is printed mainly as a curing agent of an alkyd resin and is used for a paint, since a benzoguanamine resin has a benzene machine in the structure, it is known that lightfastness is bad and a use use has restrictions. Moreover, although alkoxy ** of the melamine-formaldehyde reactant was further carried out in alcohol, such as a methanol, ethanol, propanol, an isopropanol, and a butanol, and it was made to dissolve in a solvent when a melamine formaldehyde resin did not have the solubility over an organic solvent, therefore it used as an organic-solvent system paint, since a reaction process's becoming long and alkoxy ***** were acid and performed, condensation progressed and there was [a fault like control of molecular weight comes to spread in difficulty].

[0006] Moreover, melamine resin is conventionally used mainly from the level makeup side of furniture by a high degree of hardness, high thermal resistance, and high gloss as makeup material. Since design nature is increased in recent years, the so-called post form processing which carries out post processing of the panel after hardening is carried out briskly. Moreover, while a melamine facing plate has a high degree of hardness, generating of a crack with time may be seen and the improvement is performed.

[0007] Therefore, although improvement, improvement of a crack-proof, etc. were performed in post form processability by denaturalizing to melamine resin with the compound of various copolycondensation nature, or adding various additives to it, inclusion to a resin became poor with the waterproof fall and the fall of gloss, and was not necessarily satisfactory.

[0008] Moreover, the melamine is well used as woody system adhesives combining the urea. Although excelled in the productivity by a cure rate being early as compared with the phenol system adhesives

used for the of-the-same-kind use, since the melamine system adhesives itself were lacking in lipophilic property, there was disadvantage to many [for the tar used / tar / suddenly in recent years] needle-leaf tree.

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DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] The state before applying a load in the flexibility examination of this invention is shown.

[Drawing 2] The state where the load is applied in the flexibility examination of this invention is shown.

[Description of Notations]

L0 Length before applying a load.

L Length when having applied the load.

[Translation done.]

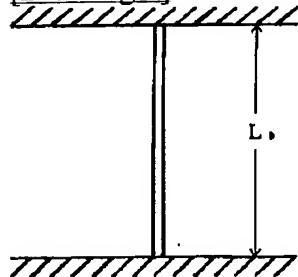
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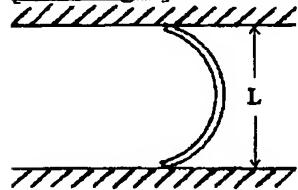
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DRAWINGS

[Drawing 1]



[Drawing 2]



[Translation done.]